A thesis titled

THE UNSTEADY STATE OPERATION OF CHEMICAL REACTORS

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ABSTRACT

The efficiency of a broad class of continuous processes operated under unsteady conditions must often be expressed as a ratio of two integrals: in chemical reactor problems this may represent the selectivity of a desired product in a complex reaction scheme. Objective functions taking this form are included in the optimal control formulation of unsteady state operation of lumped parameter continuous processes; the resultant additional necessary condition of optimality appears in a convenient form so that the complexity of the problem is only marginally increased.

The difference between the dynamic and the steady performance of continuous chemical processes is only meaningful under strictly comparable conditions. A computationally efficient procedure is developed which, without any assumptions about the form of the inputs, enables the determination of optimal continuous periodic modes of operation under comparable conditions. The proposed procedure can also be effectively used to test the optimality of a given periodic operation.

The application of the proposed procedure to chemical reactor problems under inlet control conditions indicated that in many cases the optimal steady performance can be improved by on-off periodic inputs. In particular, simultaneous increases in both the yield and selectivity of a desired product in a complex reaction scheme are attainable while using the same sources and equal average amounts of the raw materials.

SUMMARY

The potential superiority of unsteady operation over the conventional steady operation of chemical processes has come to light over the past two decades. The study presented here is concerned with the determination of optimal dynamic operation of continuous processes in general, and continuous chemical reactors in particular.

The thesis begins with a general introduction to the concepts of controlled cycling, natural oscillations, and enforced oscillations used in the unsteady state operation of chemical processes. This is followed by a discussion of conditions which enable a meaningful comparison of results from dynamic and steady operations to be made.

The second chapter opens with a survey of the literature on the enforced periodic operation of chemical reactors. Empirical methods for finding the best mode of periodic reactor operation are then examined with reference to a stirred tank reactor. There then follows a discussion on the limitations of such methods and the need for a more rigorous approach.

The remainder of the work concerns the application of optimal control theory to the rigorous determination of the best modes of unsteady state processing. After an introduction to the basic concepts of modern variational theory in chapter 3, the strongest available theorem, the Maximum Principle of Pontryagin, is stated and its qualitative utility is demonstrated through a specific unsteady state reactor problem. Chapter 3 continues

with an appraisal of the numerical difficulties inherent in the quantitative application of optimal control theory and the special nature of the objective functions which are often required. Chapter 4 is devoted entirely to this latter point: i.e. the proper inclusion of ratio-integral objectives in the application of the Maximum Principle to the unsteady state operation of continuous chemical processes.

Finally, in chapter 5 an efficient iterative procedure for the determination of optimal periodic modes of reactor operation is developed. The proposed procedure is a general one capable of handling most problems arising in the dynamic operation of lumped parameter processes. It also provides an effective means for testing the optimality of any particular periodic operation, such as one found through empirical search procedures.

An interesting conclusion drawn from the results of this work is that for many systems usually processed under steady input conditions, the optimal operating mode in fact calls for unimodal periodic on-off inputs. Such inputs are perhaps not to difficult to implement in practice.

The material in chapter 4 [A1] and parts of chapters 2 and 3 [A2] have been published.

A1. F.A.Farhad Pour,L.G.Gibilaro,"Ratio-integral objective functions in the optimal operation of chemical reactors",Chem.Engng.Sci. 1975,30,735.

A2. F.A.Farhad Pour,L.G.Gibilaro,"Continuous unsteady operation of a stirred tank reactor",Chem.Engng.Sci. 1975,30,997.

CONTENTS

CHAPTER I : INTRODUCTION

All natural phenomena are of an essentially transient nature: with the passage of time things change, edges blur and established orders decay. But this long view of the temporal scale contains within it regions where rates of change in particular observations may be either vanishingly small or subject to more or less periodic fluctuations. Such phenomena are commonplace in human experience: the human body goes through a series of states which are repeated day after day, the seasons are repeated year after year etc. The important point is that our natural surroundings behave in a dynamic manner in which time plays the major role. The basic concept in unsteady operation is time and the use that can be made of it.

1.1. Introduction

In general, continuous steady operation of chemical processes is taken as the ultimate in processing concepts, its main advantage lying in the economy of the running costs over equivalent batch operation. To offset this, however, lies the disadvantage of decreased reaction yield, the need for recycle streams and the extra separation requirements, which might not be needed in batch processing. However, in recent years growing experimental and theoretical evidence suggests that unsteady state processing could combine the economic advantages of continuous operation with the technical advantages of batch operation.

Industrial plants are composed of many intricately connected processes; consequently, dynamic operation of a particular unit could affect the performance of other units within the plant. The fluctuating outputs from a chemical reactor could adversely affect a downstream separation unit, and unsteady operation of two distillation columns in series could cause grave synchronization problems. The coordination of individual units making up a plant is a challenging but mammoth task not examined in this study, which deals with the limited problem of unsteady operation of an individual unit.

The work presented is concerned with the determination of the optimal mode of unsteady state operation of a continuous

process in general and a continuous reactor in particular. The point which distinguishes this study from the majority of the previous ones in this area, is that here the problem is posed in such a way that the unsteady operation of an already existing steady process can be considered. To this end, it is assumed that the process of interest is buffered from other units by provision of sufficient surge capacity. It is then possible to use the same sources and equal amounts of raw materials for both the steady and unsteady operation, thus enabling a direct comparison of the two modes of operation.

1.2. Dynamics of processes

Any physical process can be described by a set of inputs and outputs, the definition of the relationship between them, and the physical bounds on the variables. The process may be distillation in which case the inputs are the feed stream and the heat loads, and the outputs are the overhead and bottoms product streams. In the case of chemical reactors, the inputs are again the feed and the thermal load, and the outputs are the quantity and the quality of the products obtained.

The first step in the study of the transient behaviour of a process is the identification of the important variables and their classification into those which can be measured, controlled, or manipulated and those others which cannot. The second step is the development of a mathematical model,

using simplifying assumptions where and when necessary, to relate the input and the output variables and list the constraints. The third step is the definition of an objective function or cost criterion, and the expression of the objective as an explicit function of the process variables. In theory, once the above steps have been executed, the dynamic behaviour of the process, for any given set of inputs, can be determined and the performance measured. The inputs can then be adjusted so that their best values can be established.

The dynamics of most continuous processes can be described through a set of partial or ordinary differential equations. This study is primarily concerned with processes whose dynamics are governed by

$$\frac{d}{dt} x_i(t) = f_i(x_1(t),..,x_n(t),u_1(t),..,u_r(t),t),i=1,..,n, \qquad 1.1$$

where, the x's denote output or state variables, the u's the input or control variables, and the independent variable, t, represents time or distance. Then, if the control variables are given functions of time and the initial state of the process is specified, the course of the process may be determined by the integration of system (1.1). The performance can then be measured through a given objective function.

$$J = J(x_1(t),...,x_n(t),u_1(t),...,u_r(t),t). \qquad 1.2$$

In physical problems, the control variables, such as temperature, pressure, current, concentration, flow rate etc., cannot take on arbitrary values; nor can they be changed instantaneously. The nature of the restrictions on the inputs depends on the physics of the individual process at hand, and the speed with which the effects of a change in the inputs is reflected in the outputs. However, in the majority of situations the control constraints can be adequately expressed in the following form

$$u_j^{min} \leqslant u_j(t) \leqslant u_j^{max} \quad \text{for all } t, j=1,\ldots,r. \qquad 1.3$$

In certain cases, there may be enough power in the admissible controls to move the process to a state unacceptable from the view point of safety or reliability, for instance in temperature overshoot problems encountered during start up of a chemical reactor, or in the overheating of an engine driven at high speed in low gear: in such cases the state variables must also be bounded.

The objective function employed plays an important role in the determination of the final design of a process. In an ideal situation the objective takes into account all the individual costs which together determine the overall cost criterion. In practice however, the combined effect of all the factors which affect the performance cannot be easily expressed as a single mathematical function; and some costs

such as the social, political and ecological costs of a process are not easily measured. As a result the final design is often based on a simplified objective, or a compromise between several designs each yielding the best results for a particular objective. Even without these complications the choice of an objective in finding the best dynamic operation of chemical reactors still presents some difficulties and is considered later in this thesis.

1.3. Steady state processing

The conventional design of continuous processes is based on a stationary mode of operation in which there is no accumulation of material or energy. In steady processing the inputs and the outputs do not vary with time, and all derivatives with respect to time vanish. Distributed parameter processes are then characterised by spatial variations alone, and lumped parameter processes are described by a set of algebraic, rather than differential, equations

$$o = f_i(x_{1s},\ldots,x_{ns},u_{1s},\ldots,u_{rs}),i=1,\ldots,r. \qquad 1.4$$

The objective function, J, also becomes time invariant

$$J_s = J(x_{1s},\ldots,x_{ns},u_{1s},\ldots,u_{rs}). \qquad 1.5$$

The determination of the optimal steady operation then requires finding a constant set of acceptable controls, $u^*_{1s},\ldots,u^*_{rs}$,

which satisfy the system equations (1.4), and impart the best possible value to the objective, J_s. This is in effect an exercise in finding the greatest or least value of a function of several constrained variables.

In some cases, the optimum steady operation can be found through the classical methods of calculus or by graphical techniques. In general however, numerical procedures are called for; such procedures belong to the general field of mathematical programming. In particular, as the system equations are often nonlinear, the problem is one of nonlinear programming which has been extensively treated in the literature [1,2,3,4]. In particular, a critical review of several algorithms with the relevant flow sheets and computer programs may be found in the text by Himmelblau [2].

Under certain conditions there may be more than one steady state for a given set of constant inputs. The classic example is furnished by an exothermic reaction taking place in a continuous stirred tank reactor fitted with a cooling coil or jacket. Figure 1.1. shows the familiar heat generation against reactor temperature plot, with the heat removal lines for several cooling rates superimposed. The possibility of multiple steady states is clearly indicated by the number of intersections between the heat generation and removal curves.

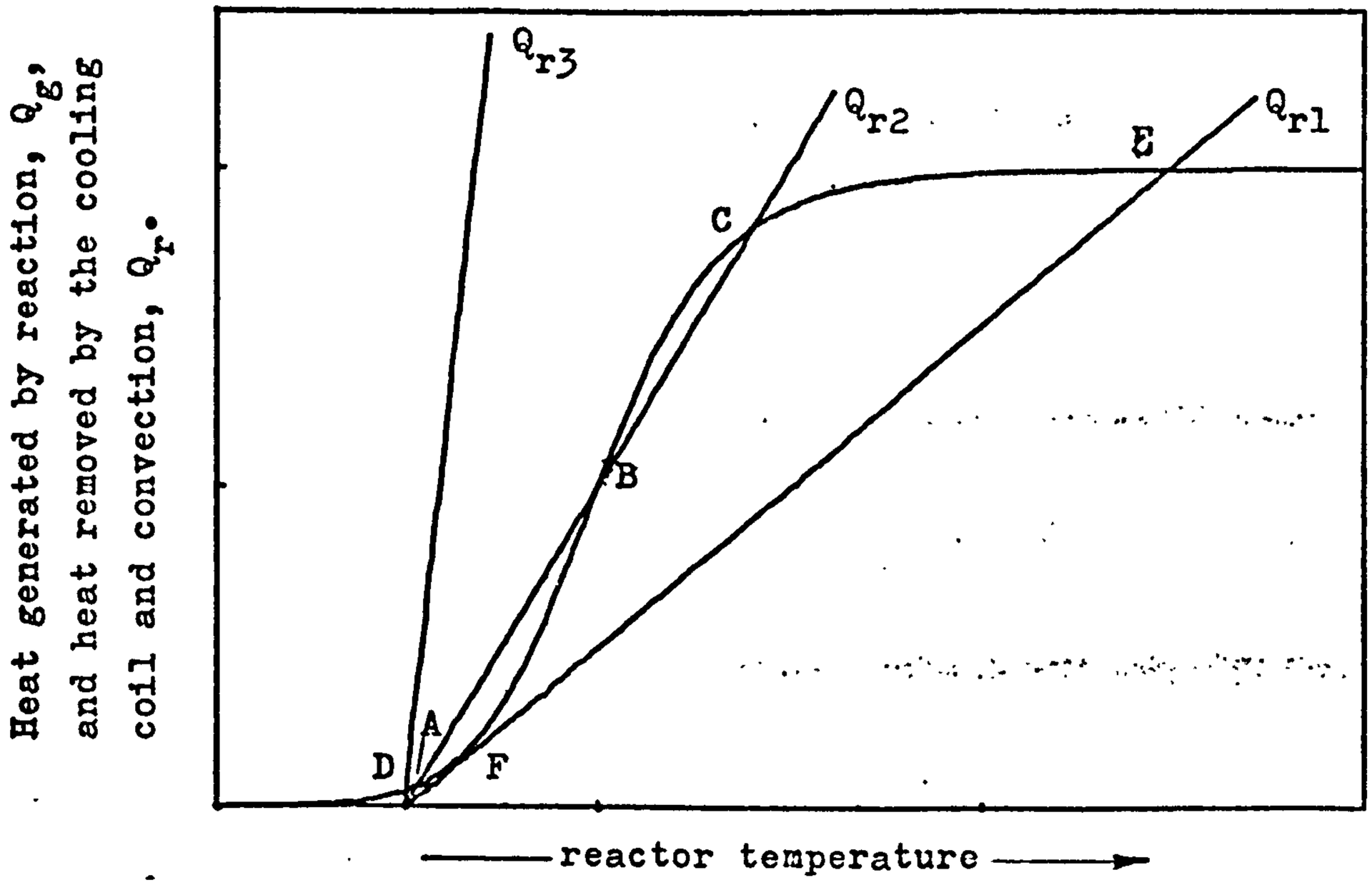


Fig.1.1. Steady states of a first order exothermic reaction in a C.S.T.R.

The essential condition for the presence of multiple steady states is the existence of a natural or induced feedback mechanism through which, the state of a process at a particular stage is linked to that of a previous stage. In a stirred tank reactor, or a tubular reactor with axial dispersion, the feedback mechanism is a natural consequence of the back mixing within the vessel. In a packed bed reactor it could arise as a result of a significant backward conduction of heat through the bed; or it could be induced through an exchange of heat between the cold ingoing and hot outgoing streams. A rather different

example of multiple steady states could arise in an adiabatic packed bed reactor in which the particles offer small mass and heat transfer resistances. In this case each individual particle could behave as a stirred tank and exhibit multiple steady states [5]. Further examples may be found in most texts on reaction engineering [6,7,8,9].

In practice some or all of the inputs to a process are prone to gradual or sudden changes, the steady state design being based on the mean value of the variable inputs. Therefore, to keep the operating levels as close to their steady design values as possible, steps must be taken to compensate for input fluctuations. This is usually achieved through the provision of surge capacity or the addition of control loops or both. The ease with which it can be accomplished depends on the stability of the process at hand.

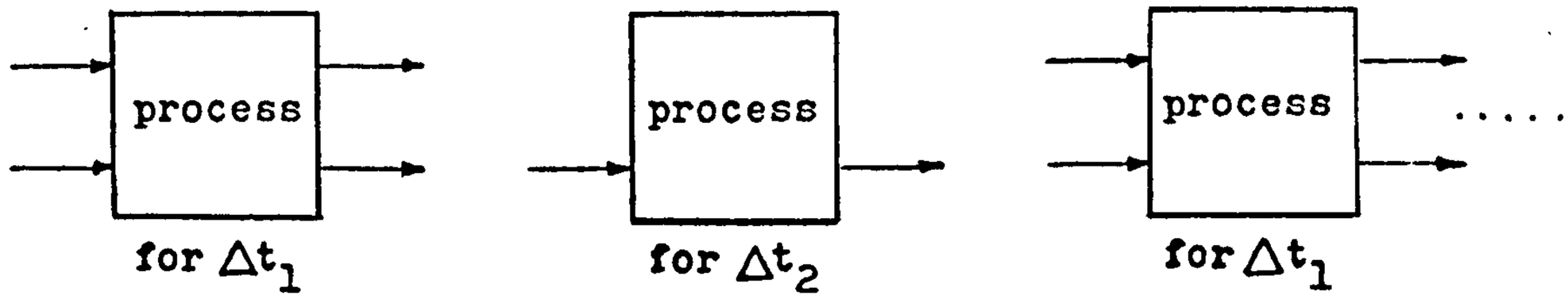
The examination of the steady behaviour of a process often yields valuable, if incomplete, insight into the understanding of stability. For the example cited in Figure 1.1, a necessary and sufficient condition for instability is a greater slope of heat generation than heat removal. Intermediate solutions, such as point B, are unstable in as far as the smallest upset in the operating temperature, causes the process to move towards point A or C. It is difficult, if not impossible, to operate the reactor at a steady state represented by point B [11,45]. A larger slope of heat removal on the other hand, provides only a necessary condition

for stability; to present a sufficient condition, the dynamic behaviour of the process in the local vicinity of a steady state must be examined. In general, the stability analysis, and the definition of the control strategies, is based on a linearised model describing the dynamics of a nonlinear process in a small region. This state of affairs can present difficulties in unsteady processing which, as will be seen later, may involve large amplitude disturbances.

1.4 Unsteady state processing

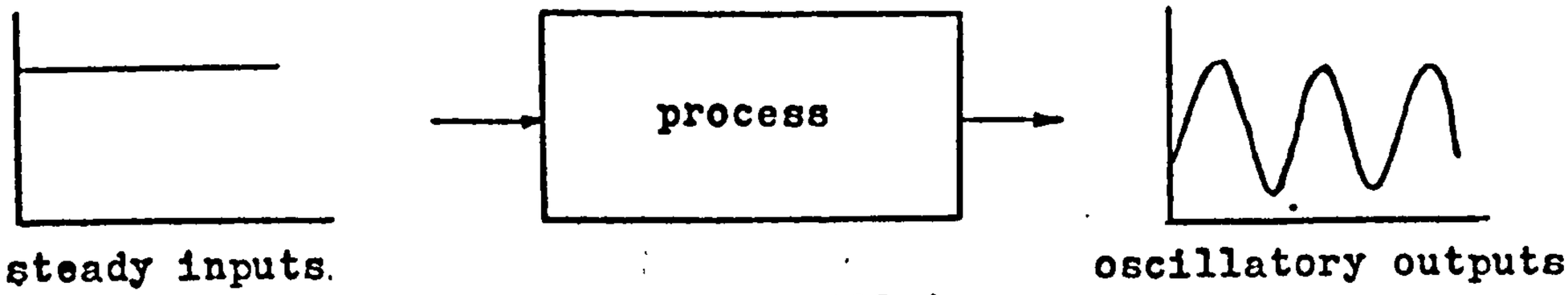
The potential superiority of unsteady processing over the conventional steady mode has been a subject of interest for some time; over the past 20 years it has been successfully applied to a variety of chemical processes. The major advances have been made for separation processes, such as distillation, extraction, crystal purification, particle separation etc. The extent of the progress made is reflected in the existance of pulsed separation units in commercial use. More recently, periodic operation of chemical reactors has been shown to result in improved conversion of raw materials.

Unsteady state processing can be accomplished in numerous ways; the common factor being that the process outputs are time variable and act over a range of values. The most widely used mode of unsteady operation is that in which some or all of the inputs and the outputs to the process are simultaneously turned on and off for fixed intervals. The term controlled cycling is often used to describe such operations.



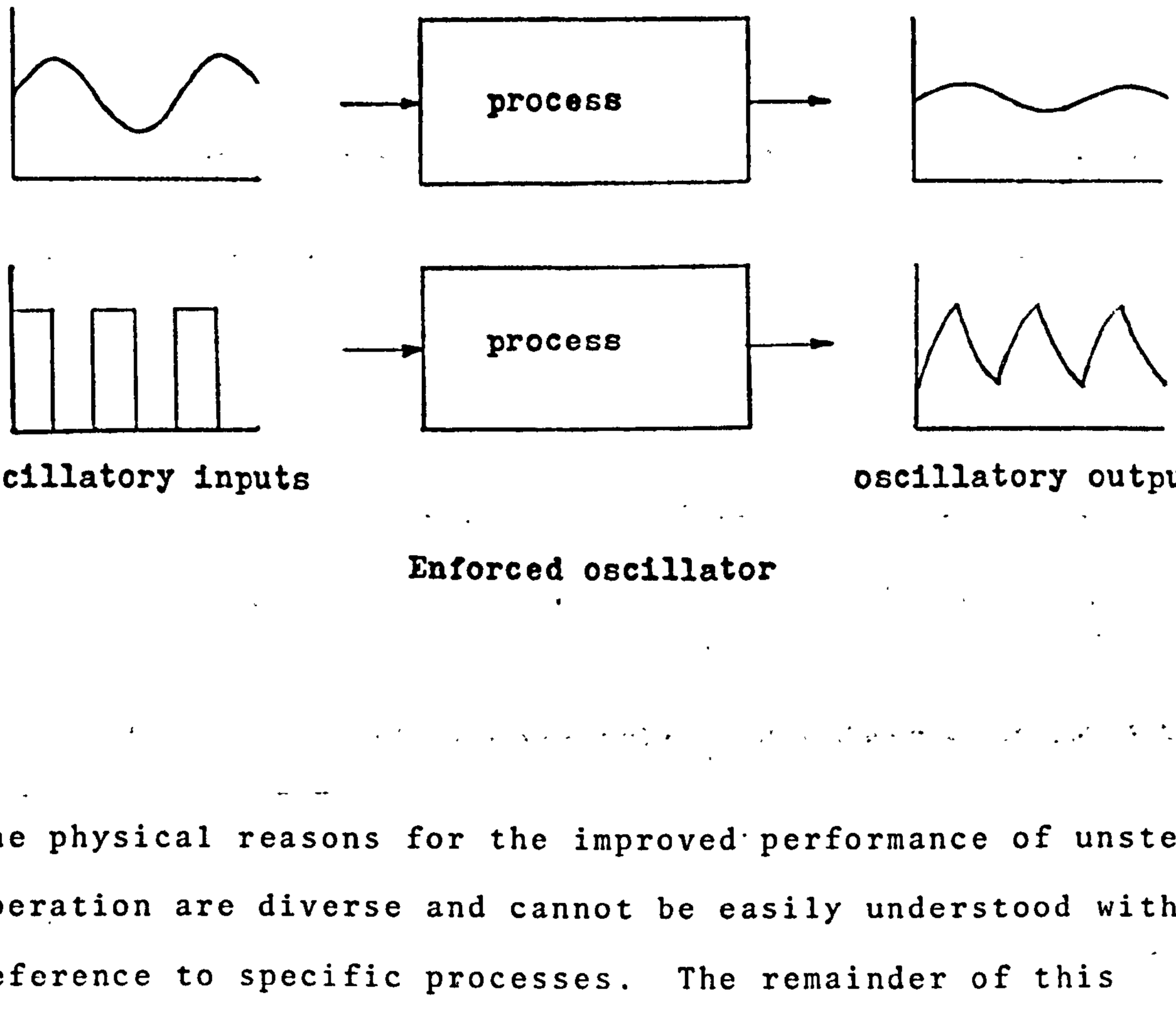
Controlled cycle operation

For a specific range of parameters, certain processes can exhibit an oscillatory behaviour even when the inputs are held steady. The design of a naturally oscillating process provides another mode of unsteady operation.



Natural oscillator

Another mode of operation, in which the outputs are not interrupted, is obtained when a controller is installed on the input side of the process, and the inputs are forced to vary either continuously or are repeatedly turned on and off for specified time intervals. In either case, the outputs assume a time variable behaviour.



Enforced oscillator

The physical reasons for the improved performance of unsteady operation are diverse and cannot be easily understood without reference to specific processes. The remainder of this chapter is devoted to a general survey of the literature, and the explanation of unsteady operation of certain illustrative processes.

1.4.1. Controlled cycling

The concept of controlled cycling was developed by Cannon [12,13] in 1956, who also guided much of the early experimental work on staged separation equipment. Such operations are characterised by the existance of intervals during which only one phase flows. For instance, a cycled distillation or gas absorption column has a vapour flow period during which the liquid remains stationary on the plates, and a liquid flow period during which no vapour flows and the liquid drains from plate to plate. In liquid-liquid extraction, coalescence

periods are added between the successive light and heavy phase flow intervals to allow phase separation. Several investigators have demonstrated that the cyclic operation of a staged process can increase both the column capacity and the overall efficiency. In distillation, capacity increases of up to three fold, and efficiency increases of around 100% are reported [14,15,16]. In extraction, the column capacity can be increased up to ten fold, and the efficiency by around 100% [17,18].

The fundamental reason for such vast improvements are best understood by comparison of a conventional and a cycled column. Consider an ideal conventional separation column with no mass transfer in the downcomers and no lateral mixing on the plates. Then, as the liquid traverses each plate, it contacts the vapour and its concentration is reduced until it reaches the downcomer and passes to the plate below without any further change in concentration. The conventional time invariant lateral concentration profiles developed on each plate are then as in Figure 1.2.

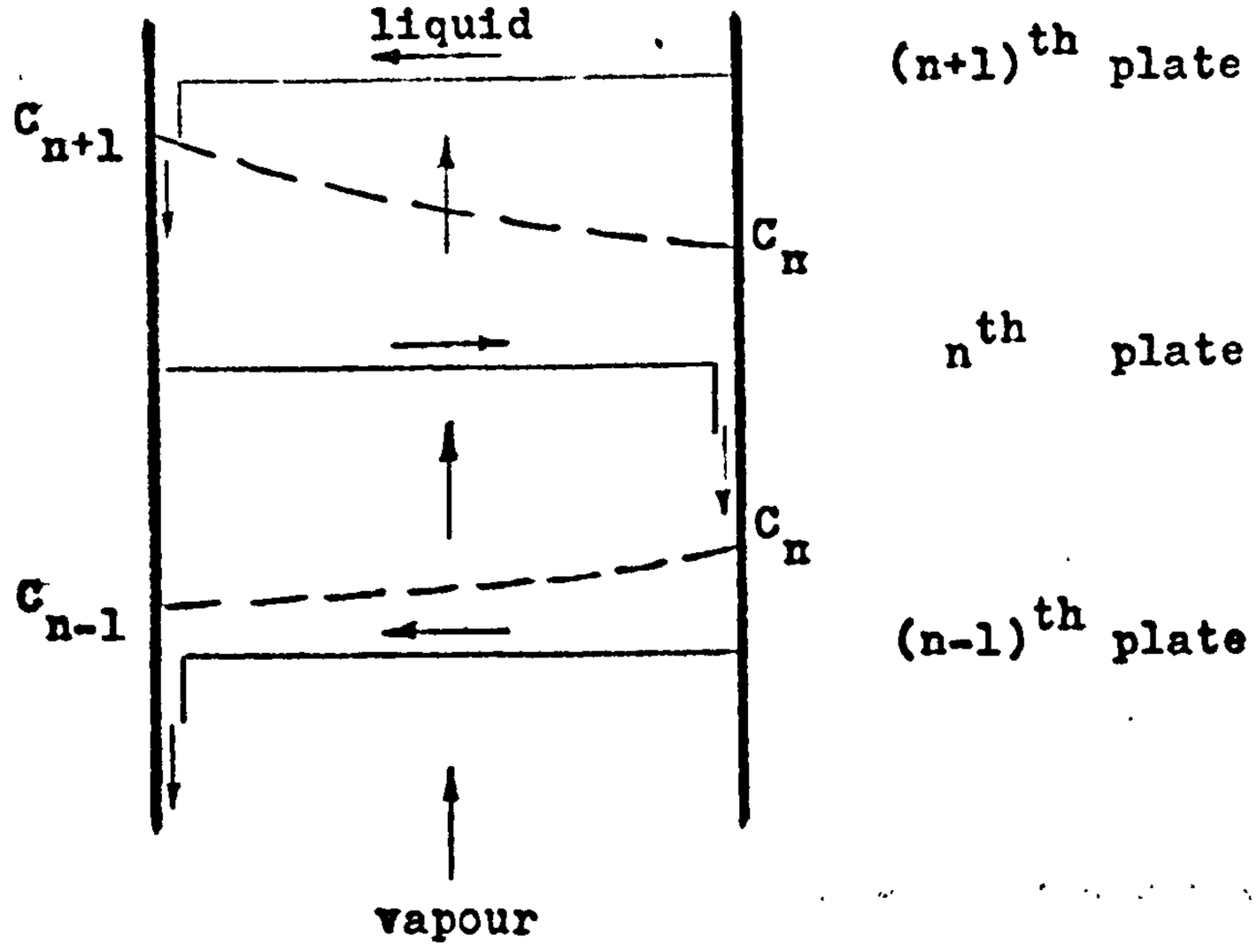


Fig.1.2. The concentration gradients in a conventional separation column.(C denotes the concentration of a key component)

Now, consider a cycled column in which all the liquid on each plate drains, with no mass transfer, to the plate below during the liquid flow periods. During vapour flow periods, the concentration of the liquid at rest on each plate is reduced until the vapour flow is shut off. Then, during the following liquid flow period, the whole content of each plate moves down to the plate below, and the vapour flow is opened again. In this case, the time variable concentration gradient on successive plates is as shown in Figure 1.3.

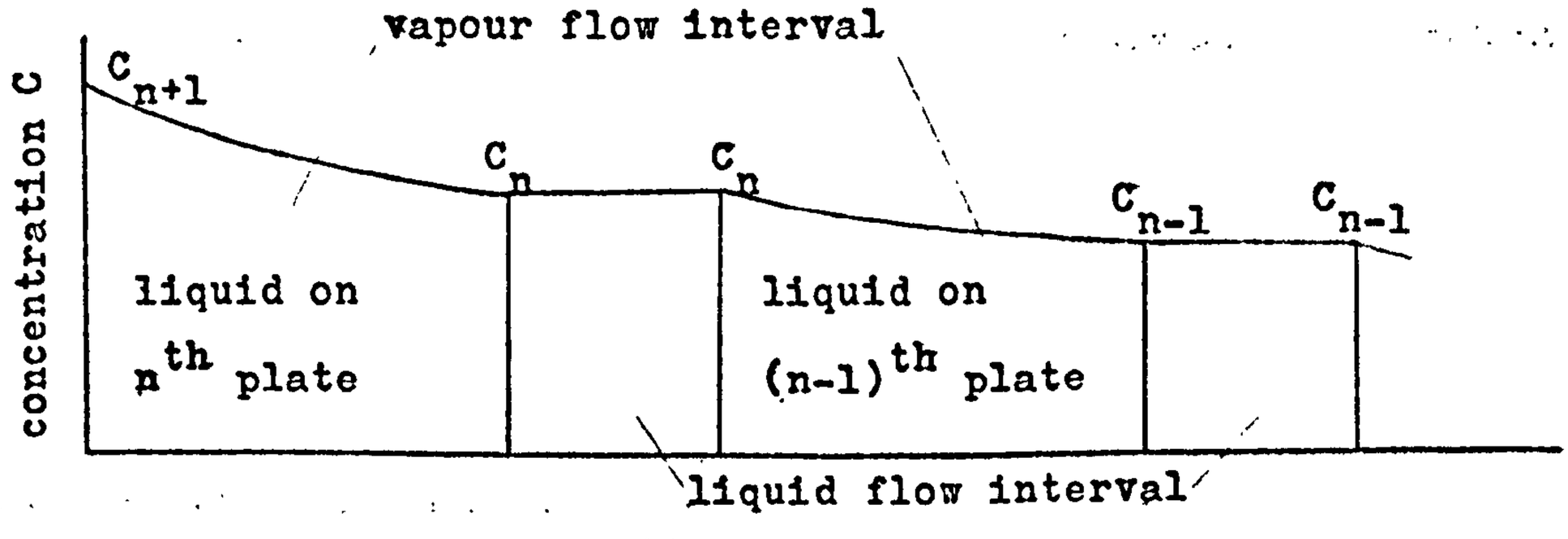


Fig.1.3. Concentration gradients in a cycled separation column.

In this case, if the vapour flow period is chosen equal to the time required for flow across a conyentional plate, and the liquid flow period is the same as the mean residence time in a conventional downcomer, the lateral concentration gradients in conventional operation are replaced by identical gradients in time. The analogy is similar to that between a batch and a plug flow reactor, with mass transfer playing the role of chemical reaction. In conventional operation, each plate resembles a continuous plug flow reactor with composition changing along its length. In cyclic operation, each plate is in effect a well mixed batch reactor with composition changing in time. The desired conventional operation is a limiting one with plug flow conditions or no lateral mixing; which is not easily achieved. In contrast, in cyclic operation lateral mixing has a desired effect; in as far as a uniform concentration on the plates reduces the effect of lateral vapour mixing. Thus, controlled cycling combines the economy of continuous

operation with the technical advantage of batch processing. This is the main reason for the improved performance of cycled apparatus.

The unsteady state operation of packed columns results in capacity increases; however, no increases in efficiency are observed [19]. In this case, the improvement is due to a change in the flow pattern through the packed column, a very flat velocity profile being the result of unsteady operation. This rather surprising development is also used in crystallisation [20] and ion exchange [21]. In crystallisation the flat velocity profile is utilized in removing the mother liquor adhering to crystal surfaces by using pure liquor to wash off the impurities. This development could also be used in adsorption or leaching, where a flat velocity profile could prove advantageous.

On the theoretical side, the analysis of controlled cycled separation processes is well advanced. McWhirter [14,22] developed the first fundamental treatment of cycled distillation columns and provided the first method for predicting the unsteady performance. Since then several other investigators have examined the cyclic operation of mass transfer units. In particular, Horn [23,24] gives a lucid treatment of the theory of multistage countercurrent separation processes.

1.4.2. Natural oscillations

Many physical, biological and chemical systems are capable of producing sustained finite amplitude oscillations even when the inputs are maintained at constant level. This phenomenon is peculiar to nonlinear processes and occurs as a direct consequence of the nonlinearities which link and couple two or more opposing characteristics. This type of behaviour has long been of interest to chemists and biologists [25,26,27] engaged in the study of chemical reactions. More recently, it has received a great deal of attention from engineers concerned with the stability and control of nonlinear systems [11,28,29].

From a conventional steady design and control point of view the possibility of such oscillatory behaviour is extremely undesirable and should be avoided at all costs. This was the consensus of opinion until ten years ago when Douglas and Rippin [30] demonstrated that sometimes an oscillating process could yield better average results than the predicted steady operation and so pioneered the use of natural oscillations as a mode of unsteady processing.

The analysis and prediction of natural oscillations has been extensively treated in the literature connected with the stability and control of chemical reactors. The basic concepts are most easily understood in terms of the feedback control of a first order exothermic reaction in an externally cooled stirred tank reactor. Under the simplifying assumption of

constant density, ρ, heat capacity, C_p, and heat of reaction, $-\Delta H$, the dynamics of this process are described by the dimensionless equations:

$$\frac{d}{d\theta} x_1 = 1 - x_1 - \alpha_1 \exp(-1/x_2) x_1 \qquad 1.6$$

$$\frac{d}{d\theta} x_2 = x_{2f} - x_2 - U(x_2 - x_{2c}) + \alpha_2 \exp(-1/x_2) x_1,$$

where

$$x_1 = A_1/A_{1f},\ x_2 = RT/E,\ x_{2f} = RT_f/E,\ x_{2c} = RT_c/E,\ \theta = tF/V,$$
$$\alpha_1 = kV/F,\ U = Ua/FC_p\rho,\ \alpha_2 = \alpha_1(-\Delta H)A_{1f}R/EC_p\rho.$$

The above process is completely bounded and it is a trivial matter to establish the upper and lower bounds on concentration, x_1, and temperature, x_2, [30,31]. The object is to operate the reactor at a given steady state, x_{1s}, x_{2s}. The control action is assumed to change the coolant flow rate such that the heat transfer coefficient, U, is adjusted in proportion to the deviation of the reactor temperature, x_2, form the desired steady value, x_{2s}. So that

$$U = U_s\ (1 + K_c(x_2 - x_{2s})\ ), \qquad 1.7$$

where K_c incorporates the gain of the controller.

The local stability of a steady state can be established

by considering a linearised version of the system equations. Introducing the deviation variables

$$y_1 = x_1 - x_{1s},\ y_2 = x_2 - x_{2s},$$

The linearised system takes the following form

$$\frac{d}{d\theta} y_1 = -(1 + \beta_1)y_1 - \beta_2 y_2, \qquad 1.8$$

$$\frac{d}{d\theta} y_2 = \frac{\alpha_2}{\alpha_1} \beta_1 y_1 - (\frac{\alpha_2}{\alpha_1} \beta_2 - \beta_3)y_2,$$

where

$$\beta_1 = \alpha_1 \exp(-1/x_{2s}),\ \beta_2 = \beta_1 x_{1s}/x_{2s}^2$$

$$\beta_3 = 1+U_s(1+K_c(x_{2s}-x_{2c})).$$

Then according to a theorem of Lyapanov [32], the stability of the nonlinear system (1.6), in a small neighbourhood of the steady state, x_{1s}, x_{2s}, is the same as that of the linearised system (1.8).

System (1.8) is stable if and only if none of its characteristic roots have positive real parts. Table 1.1. reflects the effect of the controller gain, K_c, on the roots of the linearised system, λ_1, λ_2, for a particular set of parameters.

Table 1.1.

Effect of the controller gain, K_c, on the stability of a stirred tank reactor.

$x_{2f} = x_{2c} = .035$, $\alpha_1 = \exp(25)$, $\alpha_2 = .02\alpha_1$, $x_{1s} = .5$, $x_{2s} = .4$

Feed Back	K_c	Roots	Stability
+ve	$K_c < 0$	Real $\lambda_1<0, \lambda_2>0$	Unstable
None	$K_c = 0$	Real $\lambda_1<0, \lambda_2>0$	"
-ve	$0<K_c<225$	Real $\lambda_1<0, \lambda_2>0$	"
-ve	$K_c = 225$	Real $\lambda_1 = 0, \lambda_2>0$	"
-ve	$225<k_c<45_0$	Real $\lambda_1>0, \lambda_2>0$	"
-ve	$K_c = 45_0$	Pure Imaginary	Stability Limit
-ve	$K_c>45_0$	Real $\lambda_1<0, \lambda_2<0$	Stable

Now, according to a theorem of Bendixon [32], if a phase trajectory remains inside a finite region of the phase space and does not approach a stable steady state it must itself be a closed curve (known as a limit cycle) or else approach one assymptotically. Figure 1.4 represents the result of a digital simulation of system (1.6), using a fourth order Runge-Kutta numerical integration technique. For the particular set of parameters used, all phase trajectories, irrespective of their starting point, eventually wind around the closed curve shown. The non-uniform motion of the phase point is reflected in Figure 1.5 which shows the temperature oscillations produced.

In physical terms the oscillatory behaviour is due to the coupled effect of temperature and concentration on the rate of reaction, $r = K_{1s} \exp(-E/RT)\, A_1$. As the reaction proceeds, heat is generated which in turn promotes an even faster rate of reaction. This autocatalytic phenomenon proceeds at an accelerating pace until the progressively smaller concentrations inside the tank reduce the rate of reaction. The temperature is then further reduced as the result of heat removal through the cooling coil and the reactant concentration gradually builds up to a level at which the autocatalytic phenomenon takes over and the whole sequence is repeated.

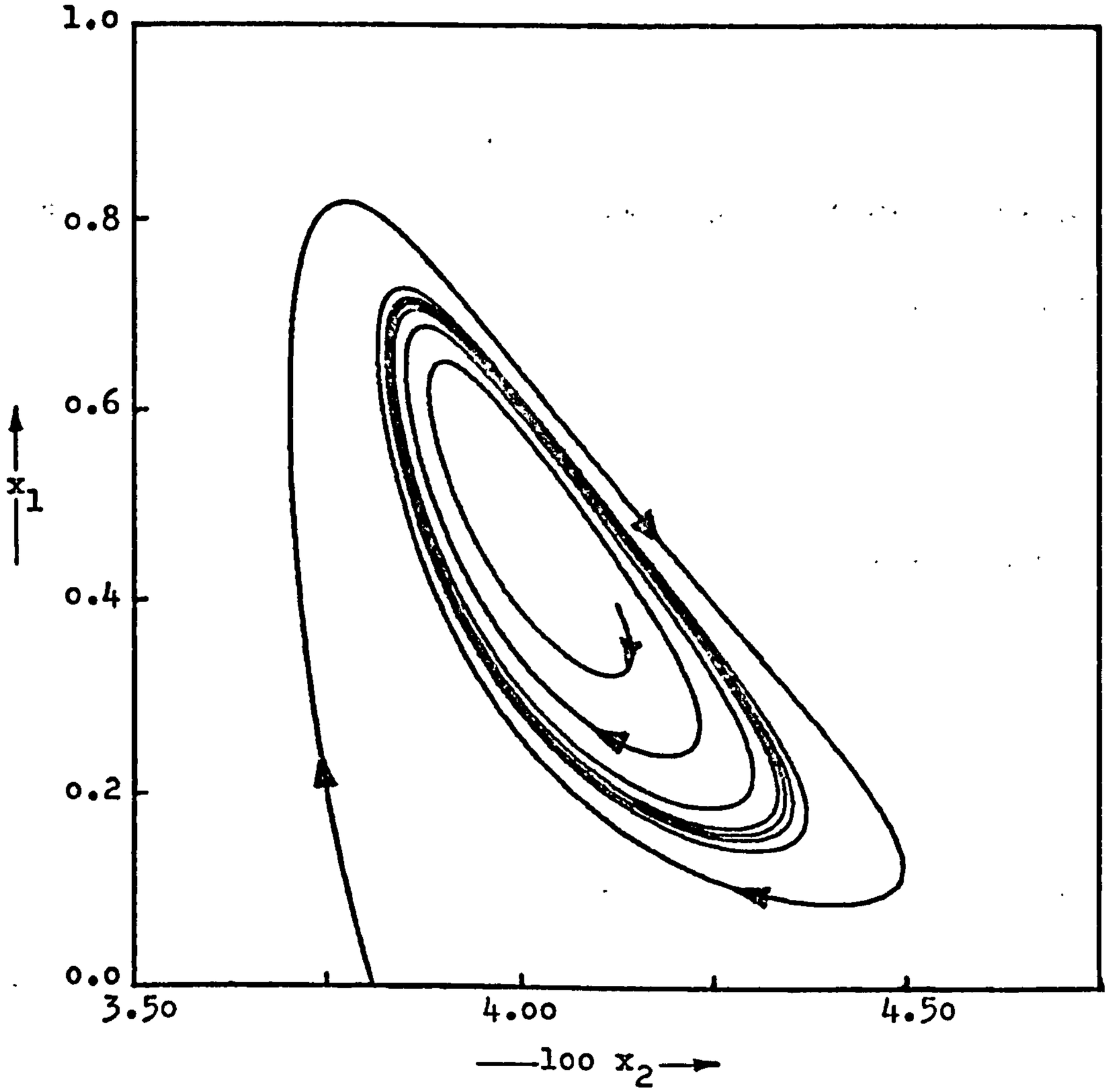


Fig.1.4. Phase portrait of an oscillating stirred tank.(K_c=400, all other parameters as in Table 1.1)

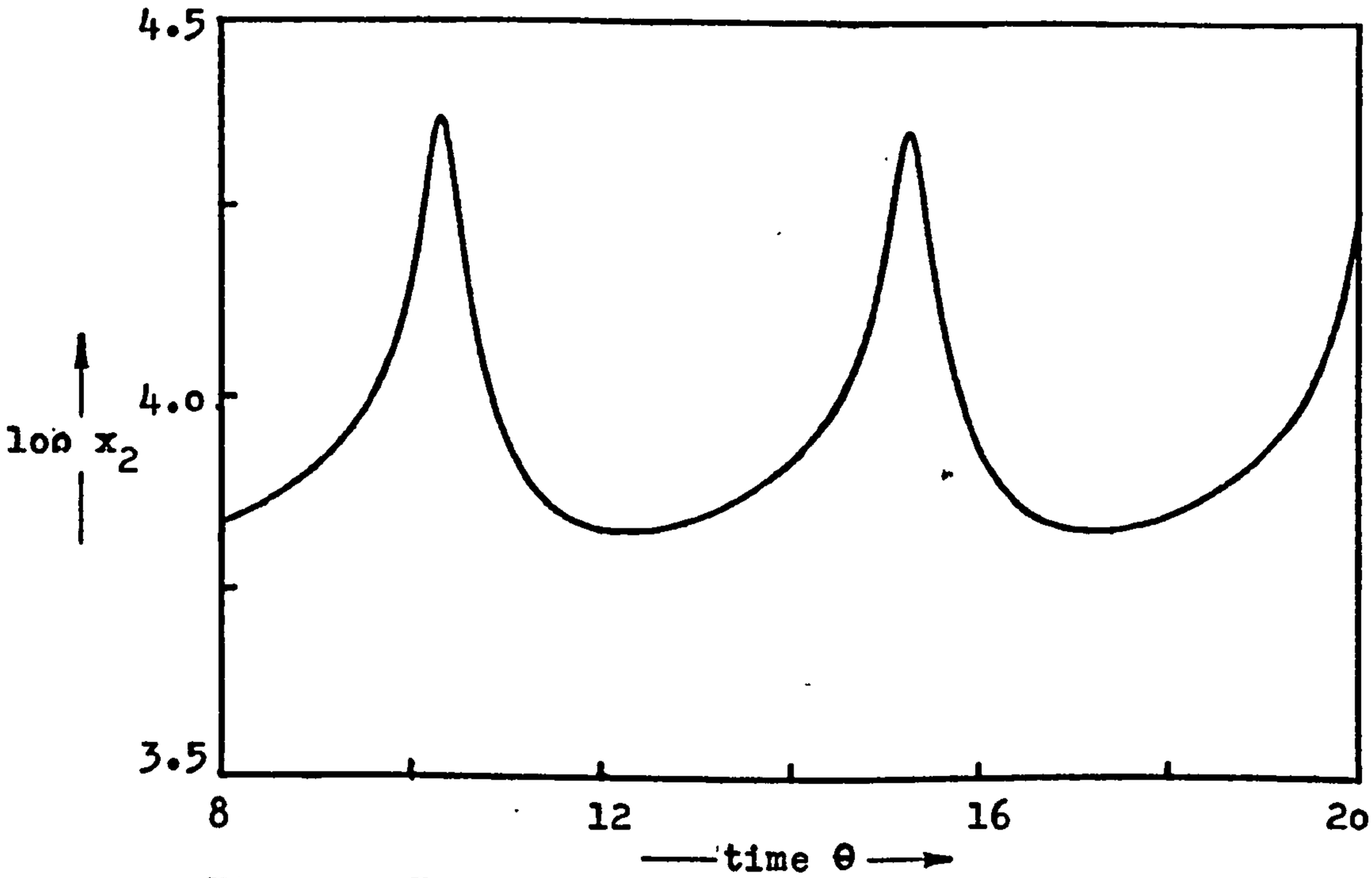


Fig.1.5. Temperature variation inside an oscillating stirred tank. (K_c=400, all other parameters as in Table 1.1)

Douglas and his co-workers have successfully utilized the fact that the average concentration from oscillating reactors are not the same as those predicted from the steady design: improvements of up to 20 % have been reported [30,31,33,37]. Analytical procedures for the prediction of the performance of oscillating processes have been developed by the same group of investigators. An interesting use of positive feedback to produce natural oscillations in an otherwise stable process has been reported by Dorawala and Douglas [33]. On the experimental side, Bush [34] has produced sustained oscillations in the successive chlorination of methyl chloride, and Baccaro et al. [35] have examined the hydrolysis of acetyl chloride.

The introductory account given above is by no means complete, many of the finer points of the analytical difficulties associated with this problem can be found in the works of Aris and Amundson [11,41,42]. Most of the more recent effort has been directed to the prediction of natural oscillations and a number of analytical [33,43],graphical [28] and numerical [29] procedures have been developed. The latest publication to date is due to Douglas [44] and deals with the design of an oscillating crystallisation unit.

1.4.3. Enforced oscillations

In practice, the range of parameter which produce natural oscillations is rather narrow and not all oscillatory processes yield improved results. Furthermore, many physical processes are by their nature incapable of producing natural oscillations. In such circumstances, the external forcing of the process inputs provides an alternative mode of unsteady state processing. The time average results from an enforced nonlinear process differ, often favourably, from that of a steady operation at the mean value of the fluctuating inputs; the magnitude of this discrepancy increases with the nonlinearity of the process. In such cases, the conventional practice of providing surge capacity and installing control loops to damp out input variations does not necessarily yield the best performance.

From a practical point of view, dynamic operations in which the inputs are subjected to regular continuous variations appear the most attractive. In general, when some or all of the inputs to a process are subjected to continuous periodic perturbations,

$$u_j(t+t_p) = u_j(t) \text{ for all } t, j=1,\ldots,r, \qquad 1.9$$

after an initial settling out interval, the outputs from the process also become periodic functions of time. An enforced periodic operation is then described by system (1.1.) Eqs. (1.9) and

$$x_i(t+t_p) = x_i(t) \text{ for all } t, i=1,\ldots,n, \qquad 1.10$$

where t_p denotes the period of oscillations. Once a periodic operation has been established, its performance can be easily measured by averaging the outputs over one complete cycle.

Naturally, a steady process may be thought of as a periodic one in which the period can take on any arbitrary value. In the same way, in the case of a naturally oscillating process, the period of the steady inputs can be arbitrarily chosen to coincide with that of the variable outputs. A batch operation which is carried out repeatedly with the same initial conditions is a trivial example of periodic operation; Horn and Lin [47] have demonstrated the similarities of some steady recycle and periodic operations.

As an illustrative example consider the effect of sinosoidal perturbation of the inlet concentrations of an isothermal C.S.T.R. in which the following second order irreversible reactions take place,

$$S_1 + S_2 \xrightarrow{r_1} S_3 \ , \ r_1 = k_{1s}A_1A_2,$$

$$S_2 + S_3 \xrightarrow{r_2} S_4 \ , \ r_2 = k_{2s}A_2A_3.$$

Figure 1.6 represents an analog simulation of the process response to identical sinosoidal variations in the input concentration of reactants S_1 and S_2. The approach to the

periodic state for various input frequencies is demonstrated in Figure 1.7; in each case, irrespective of the initial conditions within the reactor, the phase trajectory approaches a closed curve assymptotically.

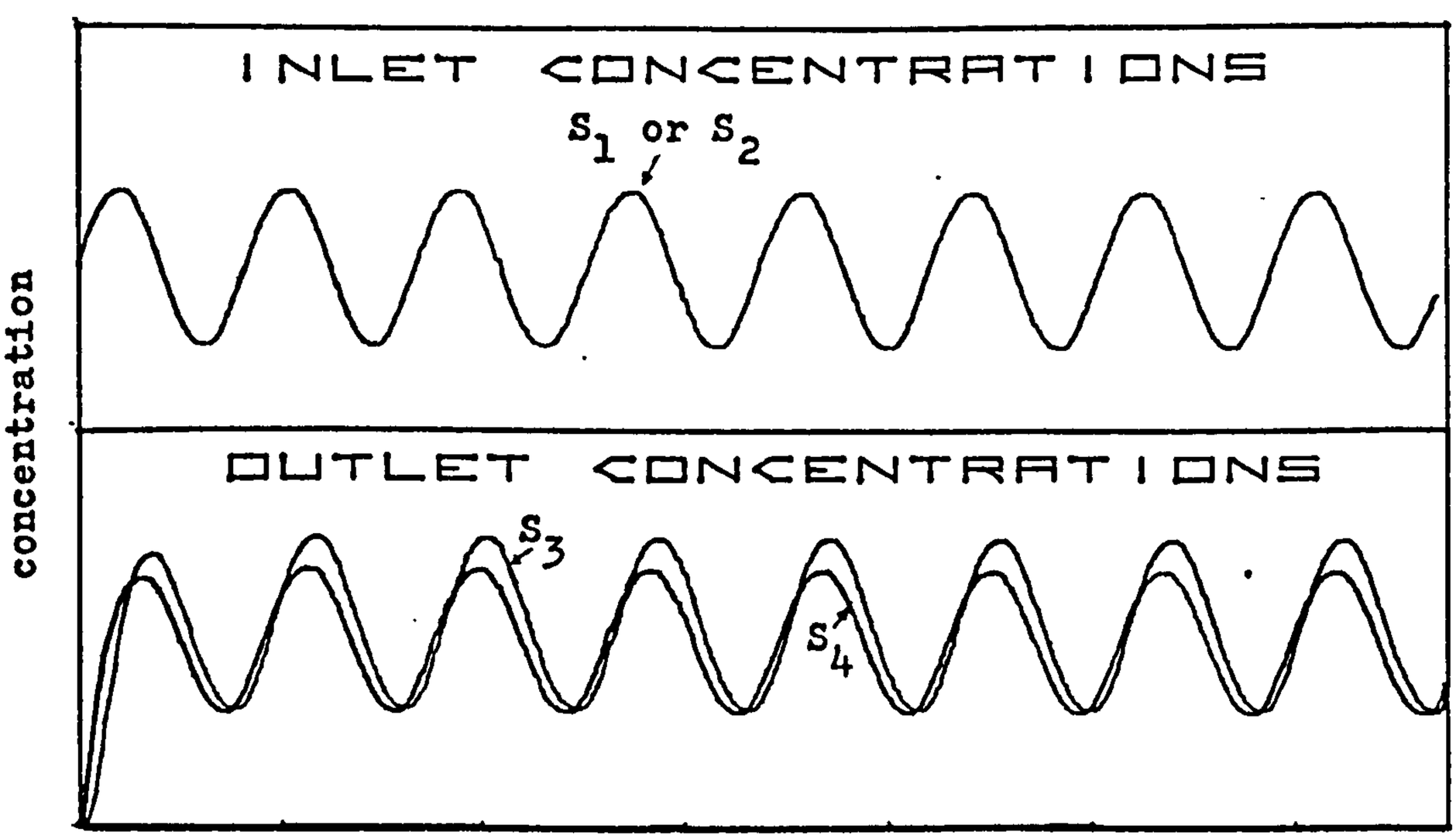


Fig.1.6. Response of an isothermal C.S.T.R. to sinosoidal inlet concentrations.($S_1+S_2 \rightarrow S_3, S_2+S_3 \rightarrow S_4$, $K_1/K_2 = 1.0$)

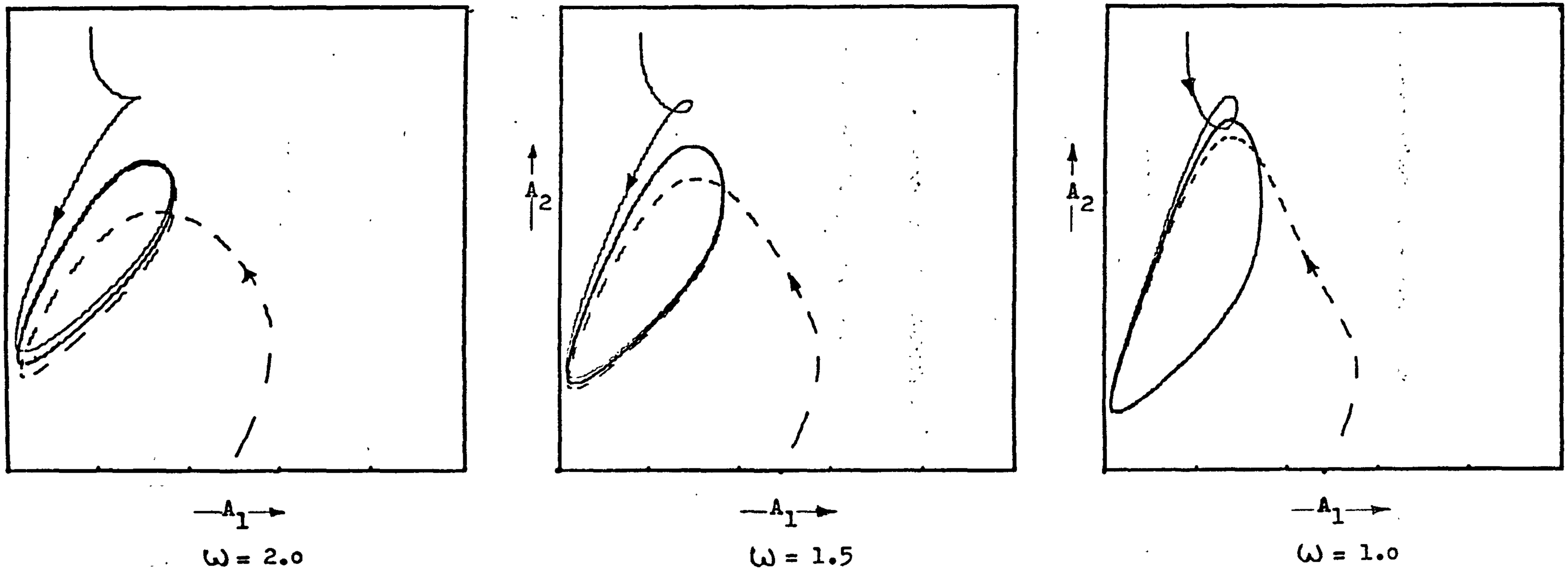


Fig.1.7. Phase portraits of an isothermal stirred tank reactor subjected to sinusoidal inlet concentration variation. $A_{if} = 0.5\, A_{1fs}\,(\,1 + \sin(\omega t)\,)$, i=1,2.

The best results may be obtained by out of phase variation of the inputs, or through simultaneous forcing of the inputs with wave forms of different frequency. Such operations could give rise to beat frequencies; the process is then capable of producing multimodal output variations which have a frequency different from that of the inputs. As a simple example consider the isothermal operation of a C.S.T.R. in which the following reactions take place

$$S_1 + S_2 \xrightarrow{r_1} S_3 \xrightarrow{r_2} S_4,$$

$$r_1 = k_{1s}A_1A_2 \ , \ r_2 = k_{2s}A_3.$$

Figure 1.8 shows the result of an analog simulation of the process for sinosoidal variations of the inlet reactant concentrations. Very slow output frequencies can be generated when the inputs vary at nearly equal rates.

Another phenomenon which could occur with enforced unsteady operation is resonance. This is a phenomenon associated with most vibrating systems and is frequently observed in everyday life: as a car decelerates vibrations are amplified at a certain speed, or when a hi-fi set plays a particular note a vase may vibrate violently or even shatter. A double-pipe steam to water heat exchanger has been shown to be capable of producing a resonance effect in the effluent water temperature, when the inlet temperature of the steam or the water are subjected to sufficiently high frequencly sinosoidal variations [39,40].

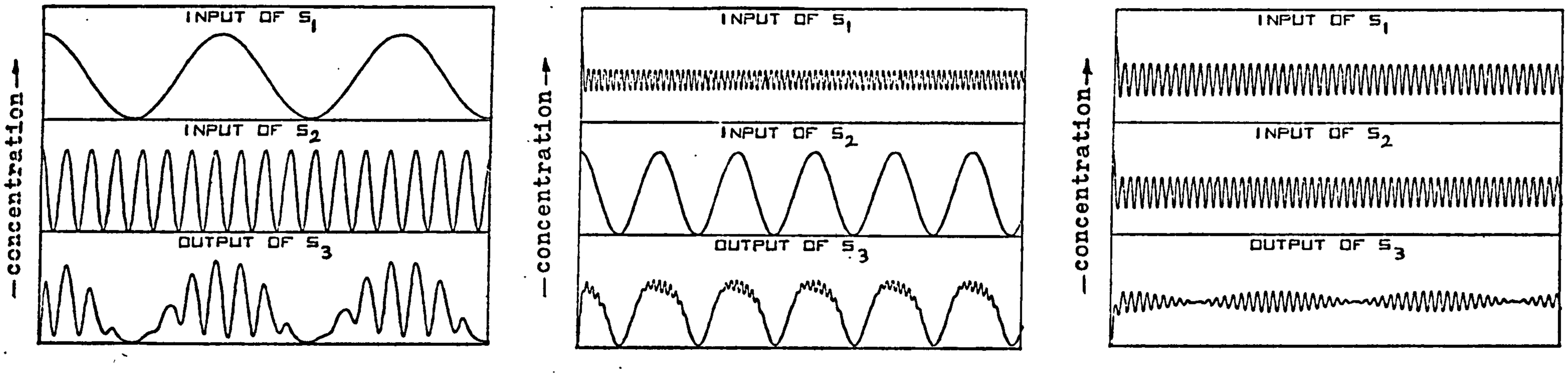


Fig.1.8. Response of an isothermal stirred tank reactor to sinosoidal inlet concentrations of different frequency.

$S_1+S_2 \rightarrow S_3 \rightarrow S_4$, $A_{if} = 0.5\, A_{1fs}(1+\sin(\omega_i t))$, $i=1,2$.

An interesting application of this resonance effect would be to force the inputs to a process capable of producing natural oscillations, with variations having the natural frequency of the process.

The enforced continuous periodic operation of many chemical processes has proved superior to conventional steady operation. Such operations can be achieved by forcing any one of the process inputs, a survey of the relevant literature as regards chemical reactors will be presented in the next chapter.

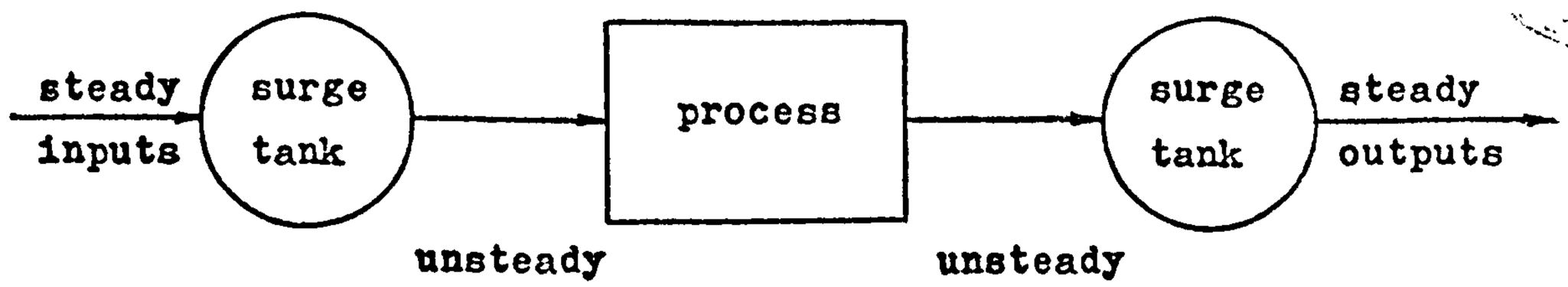
1.5. Comparison of steady and unsteady state modes of process operation

In the end, the comparison between steady and unsteady modes of processing must be based on economic grounds. In practice, unsteady operation of a particular process is advantageous only if on the average more of the desired products are produced with a running cost equal to that used in steady operation. Alternatively, dynamic processing may prove superior if the same average amount of the desired products can be produced while using smaller equipment than is necessary in steady operation. In the absence of detailed economic data, the alternative is to consider a limited objective which is suitably related to the cost of the process. The study presented here deals with such problems; however, the methods developed can easily take into account actual economic data.

The comparison of the time average results of unsteady operation with those from steady operation requires careful attention. This is because, the differences between the two modes of operation are only meaningful when comparable conditions are used - a point easily and often overlooked. In general, to obtain comparable conditions it is necessary to impose restrictions on the unsteady mode of operation. The exact nature of the constraints depends on the particular process at hand and the objective employed. There are, however, a number of guidelines which should be observed in all cases.

For instance, the time average performance should not contain any contributions from the transient intervals obtained during the start up or shut down of a process. The same control constraints should be used in the determination of the best steady and dynamic modes of operation. Furthermore, when an optimal steady operation exists, the average results of unsteady operation should not be compared with non-optimal steady results.

The primary aim of this work is the improvement of a process within an already existing steady plant. To ensure that unsteady operation of the process of interest does not upset the performance of other units within the plant, it is assumed that sufficient downstream and upstream surge capacity are available. These may already be incorporated in the steady design of the plant. In general, however, unsteady processing necessitates the introduction of additional surge capacity.



The dynamic operations envisaged must therefore require the least modification to existing plant and be easily implementable. The same sources of raw materials should be used in both the steady and unsteady modes of operation. The same average amounts of feedstock should be used in either mode, so that the cost of the raw materials remains the same. Consequently, the unsteady modes of operation must satisfy the following conditions:

I) The variable control parameters in dynamic operation do not at any time exceed the corresponding steady level.

II) The same average control efforts are used in both the dynamic and steady modes of operation.

The first condition refers to the quality of the raw materials used and the second to the quantity. For instance, if the control variable is a fuel, the first condition implies that the same grade of fuel should be used in both the dynamic and steady modes of operation, while the second implies that equal time average amounts of the fuel should be used in either case.

The remaining chapters of this thesis are concerned with the step by step identification and solution of the problems encountered in the determination of the optimal unsteady state operation of continuous processes. Although particular emphasis is put on chemical reactors, many of the arguments and results obtained are applicable to other continuous processes.

CHAPTER 2

ENFORCED PERIODIC OPERATION OF CHEMICAL REACTORS : AN EMPIRICAL APPROACH.

In recent years numerous investigations have revealed that unsteady operation of chemical processes often proves superior to steady operation. In such cases, the conventional design and control criteria do not correspond to the best performance. The field of reaction engineering is an area where unsteady processing can display significant advantages.

Over the past decade several publications in the chemical engineering and optimisation literature have examined the dynamic operation of chemical reactors. Two distinct processing concepts have been employed. One of these, the design of naturally oscillating reactors, is not pursued any further here as, in general, the range of parameters which produce a superior average performance is rather narrow and very little control of the self excited output oscillations is possible; furthermore, many reaction systems are inherently incapable of producing natural oscillations.

Instead we will concern ourselves with dynamic operations accomplished by the external forcing of process inputs.
In this chapter we examine the empirical approach to the determination of optimal periodic input profiles. Particular emphasis is placed on a point often overlooked, namely the definition of constraints under which the steady and unsteady operations can be justifiably compared.

Comprehensive coverage of the literature on all aspects of reaction engineering may be found in the annual reviews of Kinetics and Reaction Engineering published by Industrial and

Engineering Chemistry. The steady design of chemical reactors is a well established procedure which has been the subject of numerous textbooks and publications. The dynamics of chemical reactors have also been extensively studied in connection with their stability, control and optimum start up conditions. The design of chemical oscillators as a means for dynamic operation was briefly discussed in section 1.4.2, where references to the previously published work can be found. The survey presented below deals specifically with enforced unsteady state processing of chemical reactors.

2.1. Literature Survey: enforced periodic operation of chemical reactors

In the conventional design of a chemical reactor provisions are made to damp out input variations caused by upstream fluctuations from other processing units and the external sources which supply the reactor; paradoxically it may be that leaving these input variations unchecked, or even amplifying them, will result in an improved performance.

Douglas and Rippin [30] showed that in the isothermal operation of a stirred tank reactor with the second order reaction

$$2S_1 \xrightarrow{r_1} S_2, r_1 = k_{1s} A_1^2,$$

sinosoidal variations of the inlet concentration, A_{1f}, about its steady design value, A_{1fs},

$$A_{1f} = A_{1fs}(1+a \sin(w_a t)),$$

resulted in a higher average degree of conversion than that attained with a steady input at A_{1fs}. The magnitude of the improvements however were small, being about 0.02% with a 10% amplitude (a=0.1) variation and rising to just 0.06% at double this variation.

Similar fluctuations in the volumetric flow, F, through the reactor

$$F = F_s(1+b \sin(w_b t)),$$

did not yield an improved performance. However, when flow variations were coupled with fluctuations in feed composition, the periodic operation was once again superior to a steady operation at F_s and A_{1fs}. In this case, the magnitude of the improvement was dependent on the relative values of the frequencies, w_a and w_b, and the phase lag between the disturbances. The maximum 0.8% improvement in conversion with 10% amplilude (a=b=0.1) fluctuations occured with $w_a=w_b$ and a 180^o phase lag.

Following this early (1966) publication Douglas [36] and later Douglas and Gaitonde [31] and Ritter and Douglas [37] applied the standard methods of nonlinear mechanics [32] to the determination of the frequency response of a nonlinear stirred tank reactor and presented approximate analytical procedures with sinosoidal inputs. The detailed mathematics of these

methods, although by no means complex, is exceptionally lengthy and tedious and will not be repeated here.

Lannus and Kershenbaum [38] examined sinosoidal feed composition variation of an isothermal tubular reactor with second order kinetics. Their numerical calculations, using the isothermal axial dispersion model with closed boundary conditions, revealed that the small improvement in conversion was enhanced by the degree of mixing inside the vessel: for the two limiting cases of plug and well mixed flow conditions the improvements were of the order of 0.02% and 0.12% respectively.

The literature cited so far deals with isothermal conditions. It should be possible to observe much larger improvements for nonisothermal conditions as the inclusion of heat effects introduces an exponentional nonlinearity into the process. This effect is reflected in a study by Dorawala and Douglas [33] who examined a stirred tank reactor with the exothermic reaction schemes

$$2S_1 \xrightarrow{r_1} S_2 \xrightarrow{r_2} S_3,$$

$$r_1=k_1\exp(-E_1/RT)A_1^2, r_2=k_2\exp(-E_2/RT)A_2,$$

and

$$2S_1 \xrightarrow{r_1} S_2,\ S_1 \xrightarrow{r_2} S_3,$$

$$r_1=k_1\exp(-E_1/RT)A_1^2, r_2=k_2\exp(-E_2/RT)A_1.$$

Under isothermal conditions, the maximum improvement in the yield of the desired product, S_2, with a 10% amplitude sinosoidal flow variation were of the order of 0.02% and 0.1% for the consecutive and parallel reaction schemes respectively. Under nonisothermal conditions the same flow variations gave improvements of the order of 0.1% and 2.0% respectively. More markedly when a 10% amplitude variation in the inlet temperature was examined, improvements of up to 15% occured for the parallel reaction.

It should be noted that random fluctuations in the streams which form the inputs to the reactor are unlikely to have the desired form, amplitude or frequency. So that the input variations must in general be artificially induced, amplified or modulated. Furthermore, the periodic performance should be measured against the best steady operation under strictly comparable conditions: in many cases these do not correspond to the mean values of the periodic inputs but to their maximum level. Renken [46] tackled this problem realistically by considering periodic switching of the input concentrations between zero and the corresponding optimum steady level. The results demonstrated that for an isothermal stirred tank reactor with the reaction scheme

$$S_1 + S_2 \xrightarrow{r_1} S_3, r_1 = k_{1s} A_1 A_2,$$

$$S_2 + S_3 \xrightarrow{r_2} S_4, r_2 = k_{2s} A_2 A_3,$$

improvements in both the yield and selectivity of the desired product, S_3, were possible with feed stock concentrations no higher than those used in the steady state.

The enforced periodic operation of a nonisothermal stirred tank reactor with the exothermic reactions

$$S_1 \xrightarrow{r_1} S_2, r_1 = k_1 \exp(-E_1/RT) A_1^v, \text{ a } v^{th} \text{ order reaction}$$

$$S_1 \xrightarrow{r_2} S_3, r_2 = k_2 \exp(-E_2/RT) A_1, \text{ a 1st order reaction} \qquad 2.1$$

has been the subject of a number of studies. For this reaction scheme, it can be easily demonstrated (see Appendix 2) that provided

$$vE_2/E_1 > 1, \qquad 2.2$$

There is an optimum steady temperature corresponding to the maximum yield of the desired product, S_2; otherwise the best steady yield is obtained with the highest possible temperature. Thus, if condition (2.2) is satisfied the comparison of periodic and steady operations presents no difficulty.

In a classic publication, Horn and Lin [47] presented a fundamental approach to periodic processing and discussed its relationship with the other conventional modes of operation. Examining reactions (2.1) in a stirred tank reactor, they demonstrated that under the idealised assumption of perfect control over the reactor temperature and provided that

$$\frac{1}{v} < \frac{E_2}{E_1} < 1, \qquad 2.3$$

the periodic switching of the reactor temperature between its limits was superior to the optimal steady operation. The 20% maximum improvement in the yield of the desired product, S_2, was achieved when the switching frequency was as high as possible. Although very fast switching of the temperature is not a practical proposition, this limiting case of periodic operation is of some theoretical interest and will be discussed in more detail in a later section.

Bailey, Horn and Lin [48] have since examined the effect of including heat transfer resistance of the stirred tank, by assuming perfect control over the net heat flux to the reactor, rather than its temperature. In this case, the cyclic switching of the heat flux between its limits gave a superior performance. However, the maximum improvements were obtained not with an extremely high switching frequency, but with a finite one.

Matsubara et al. [49,50] have examined reactions (2.1) analytically and confirmed much of the results previously obtained by numerical calculations; they have further extended the analysis to the consecutive reactions

$$S_1 \xrightarrow{r_1} S_2 \xrightarrow{r_2} S_3,$$

and the reversible reactions

$$S_1 \underset{r_2}{\overset{r_1}{\rightleftharpoons}} S_2,$$

with r_1 and r_2 as given in Eqs. (2.1). Some analytical results concerning a practical situation where the reactor temperature is controlled by adjusting the coolant flow rate are also reported by the same authors.

The unsteady processing of catalytic packed bed reactors promises many interesting applications. In some packed beds the reaction components are adsorbed on the catalyst surface at different rates. The reactor is then capable of chromatographic separation, which could decrease the backward rate of reversible reactions by separating two or more of the products formed.

To employ this effect advantageously, the reactants must be injected in some pulse-like fashion into a diluent or carrier gas stream. Since 1961 several patents [51,52,53], experimental investigations [54,55,56] and theoretical studies [57,58,59] have revealed that such operations can significantly improve the conversion achieved in an isothermal packed bed reactor. In some cases conversions higher than the equilibrium conversions were obtained. These studies were confined to situations where no interaction between successive pulses took place. This could be achieved by sufficient spacing of the pulses and the use of a large flow of diluent.

However it is often undesirable to have a large flow of the diluent through the bed. In such cases, the comparison between the steady and pulsed operation of the reactor should be made under the constraint of equal average inlet conditions, and the interaction between successive pulses cannot be ignored.

This problem has been tackled by Gore [60] using a mathematical model of the isothermal chromatographic reactor. Substantial improvements over the steady conversions were reported, the magnitude of the improvements being favoured by: fast reaction rates, impulse like feed pulses, and input frequencies which give effective separation without excessive interaction between successive pulses.

The unsteady operation of a fixed bed reactor with no chromatographic effects can also be superior to the conventional mode of operation. With a complicated reaction scheme involving many components, the most important property of a catalyst could be its selectivity. Using a mathematical model of an isothermal catalyst pellet with no internal mass transfer resistance, Horn and Bailey [61] obtained significant improvement in the selectivity of a desired product for a simple heterogenous reaction scheme where the concentration of reactant in the gas surrounding the catalyst was rapidly switched between zero and a fixed upper limit.

The same authors [62] considered the high frequency switching of the inlet concentration to an isothermal fixed bed reactor operating under plug flow conditions. In this case the magnitude of the improvements in selectivity were smaller than that for a single particle with perfect control of the bulk phase surrounding it.

Bailey, Horn and Lin [48] have examined the effect of including the mass transfer resistance in a single catalyst pellet

by lumping all the resistances into a stagnant boundary layer around the active surface. The simulation studies revealed that in this case an optimum switching frequency existed, and rapid switching did not correspond to maximum selectivity.

The physical reasons for improvements when no chromatographic effects are present is more difficult to ascertain. In broad terms, it must be attributed to the concentration variations within the pellets becoming out of phase as the result of the different resistances offered to the various species. This problem is at present subject of research at Imperial College, London University [64]. The experimental results obtained for the hydrogenation of butadiene in an isothermal catalytic fixed bed reactor have given up to 30% improvement in the selectivity of a desired intermediate product when the inlet concentrations are varied as a symmetrical square wave.

In the most recent publication to date, Renken, Muller and Wandrey [63] have examined the catalytic oxidation of ethylene. The experimental results reported demonstrate that periodic switching of the reactant concentration can significantly increase the yield of the desired ethylene oxide. Periodic operation is also shown to be capable of preventing the ignition of the reactor caused by the high heats of reaction of the undesired reactions So that conversions not possible in the steady state can be obtained with a suitable periodic operation.

2.2. Periodic process operation: an empirical approach

Consider a process whose dynamic behaviour is governed by the system of differential equations:

$$\frac{dx_i}{dt} = f_i(x_1,\ldots,x_n,u_1,\ldots,u_r), i=1,\ldots,n \qquad 2.4$$

where the n output or state trajectories, $x_1(t),\ldots,x_n(t)$, are determined by the choice of the r input or control histories, $u_1(t),\ldots,u_r(t)$.

An alternative to the conventional steady state operation is to employ time variable dynamic inputs. From a practical point of view, input variations which are regularly repeated appear the most attractive. If one or more of the process inputs are subjected to piecewise continuous perturbations of some kind,

$$u_j(t) = u_j(t + t_p) \text{ for any } t,\ j=1,\ldots,r, \qquad 2.5$$

subject to any physical constraints present,

$$u_j^{min} \leqslant u_j(t) \leqslant u_j^{max},\ o \leqslant t \leqslant t_p,\ j=1,\ldots,r, \qquad 2.6$$

Usually after an initial settling out period, which is discounted, the outputs from the process also proceed to cycle regularly

$$x_i(t) = x_i(t + t_p) \text{ for any } t,i=1,\ldots,n. \qquad 2.7$$

The time average performance of the periodic operation can then be measured through a given objective function

$$J = \frac{1}{t_p}\int_{t}^{t+t_p} f_o(x_1,\ldots,x_n,u_1,\ldots,u_r)dt, \qquad 2.8$$

where $f_o(.,.)$ is some measure of the instantaneous profit.

The problem is then to determine the period t_p and the periodic input profiles, $u_j(t), j=1,\ldots,r$, such that the objective, J, assumes its best possible value. This is a problem considered in the calculus of variation; its complete solution, as will be seen later reduces to that of a complicated two-point boundary-value problem and is not easily accomplished.

However, if each control variable, $u_j(t)$, is replaced by a periodic expression containing several adjustable parameters, the objective, J, can be viewed as a function of these and, for a given set of parameters, can be evaluated by the forward integration of the system equations (2.4) until the periodicity condition (2.7) is satisfied. The best values of the input parameters can therefore be found through a search procedure for the extrema of a function of these constrained parameters and the variational problem is reduced to the much simpler one of mathematical programming.

In this way suboptimal modes of periodic operation can be found with relative ease. Computationally, the most difficult part is the repeated solution of an initial value problem

which, although time consuming, does not present a major obstacle. Naturally, as the number of adjustable input parameters are increased the true optimum input profiles are approximated more closely. However, the search procedure can become increasingly time consuming.

This approach is an empirical one, in so far as the parametric input waveforms are chosen beforehand. It is, however, an attractive first step since the input waveforms can be selected with the physical limitations of the process, and the constraints necessary for a meaningful comparison of the different modes of operation, in mind.

2.3. Continuous periodic operation of chemical reactors

In looking at the different ways of operating a given chemical reactor the objective function should take into account the cost of feedstocks, the value of the products formed and the ease with which they can be separated. In the absence of detailed economic data the performance of a chemical reactor is usually measured in terms of either the overall yield of a desired product , η,

$$\bar{\eta} = \frac{\text{amount of the desired product formed over a given time interval T}}{\text{amount of key reactant fed to the reactor over a given time interval T}},$$

or the overall selectivity of a desired product which may be defined as

$$\sigma = \frac{\text{amount of the desired product formed over a given time interval T}}{\text{amount of a key reactant converted over a given time interval T}} .$$

In situations where the products can be easily separated and the by-products are of some monetary value, the overall yield has the greater economic significance. However, when the side reactions are particularly undesirable, the by-products are of little value and product separation difficult or expensive, the overall selectivity assumes an increasingly important role.

In a continous flow reactor with complicated reactions, the operation with maximum selectivity often corresponds to neglegible production rate of the desired as well as the undesired products. This is because, the residence time and consequently the conversion of reactants, must be so small that there is no time for the destruction of the desired product through undesirable side reactions. In contrast, the operation with maximum yield often carries the penalty of having large outputs of the undesired products associated with the maximum production rate of the desired one. In practice therefore, the performance must be measured in terms of a compromise between the overall yield and selectivity of a desired product.

The periodic operations envisaged here are obtained by the periodic forcing of the reactor inlet concentrations subject to the following constraints.

I. The periodic operation is a continous one in which the flow rate is maintained at a fixed value during the entire operation.

II. The reactants are available from external sources with fixed concentrations which cannot be exceeded in periodic operation.

III. The same average amount of the reactants reach the reactor in the periodic and steady modes of operation.

These conditions are in keeping with remarks made in Section 1.5 and justify comparison between the various modes of operation. To satisfy conditions I to III, it is assumed that an unlimited supply of diluent is available so that the inlet concentrations can be diluted down from their source levels in any given form. Consequently, the time average concentrations in periodic operation are always smaller than in the steady state. The volumetric flow rate is then correspondingly increased so that the average amount of reactants reaching the reactor in unit time is maintained constant for all modes of operation.

As the improvements with unsteady operation are due to the nonlinearity of a reaction system, it could be argued that the largest improvements will be obtained by switching the inlet concentrations between the steady value and zero which employs the full amplitude of the variation possible and so forces the reactor as far away from the linear region as possible.

2.3.1. The isothermal stirred tank reactor

In the majority of previous work on periodic operation of isothermal stirred tank reactors the average periodic performance is measured against that from a steady operation at the mean value of the periodic input. In situations where the best steady results are obtained with the highest possible concentration this comparison is not valid. In such cases, it should be made with a steady operation at the maximum value of the periodic inputs and not at their mean. This is the underlying reason for the introduction of condition II above.

Renken [46], using similar constraints to conditions I to III, examined the general consecutive competing reactions,

$$S_1 + S_2 \xrightarrow{r_1} S_3 \;,\; r_1 = k_{1s}\, A_1.A_2$$

$$S_2 + S_3 \xrightarrow{r_2} S_4 \;,\; r_2 = k_{2s}\, A_2.A_3 \qquad 2.9$$

and obtained some improvements in yield and selectivity of the deisred product, S_3, when the feed stream concentrations were forced as a symmetrical square wave. We will now show that for this case further improvements in performance can be obtained by means of a small modification to the periodic input functions and then examine other on-off variations.

For fixed inlet concentrations, A_{1fs},A_{2fs},it is an easy matter to show that for the above reaction scheme there is an optimum mean residence time corresponding to the maximum yield of the desired product, S_3. The selectivity, however, falls from unity at small conversions to zero at full conversion. So that a steady operation with maximum selectivity is a trivial one with negligible production rate. Figures 2.1 and 2.2 demonstrate the effect of conversion on the steady state yield and selectivity of the desired product, S_3, for several values of the rate constants K_1 and K_2.

Referring all concentrations to the optimum steady state input concentration of reactant S_1,A_{1fs}, the flow rate to the optimum steady flow rate, F_s, and time to the optimum steady mean residence time, τ_s, the process is described by the following dimensionless equations

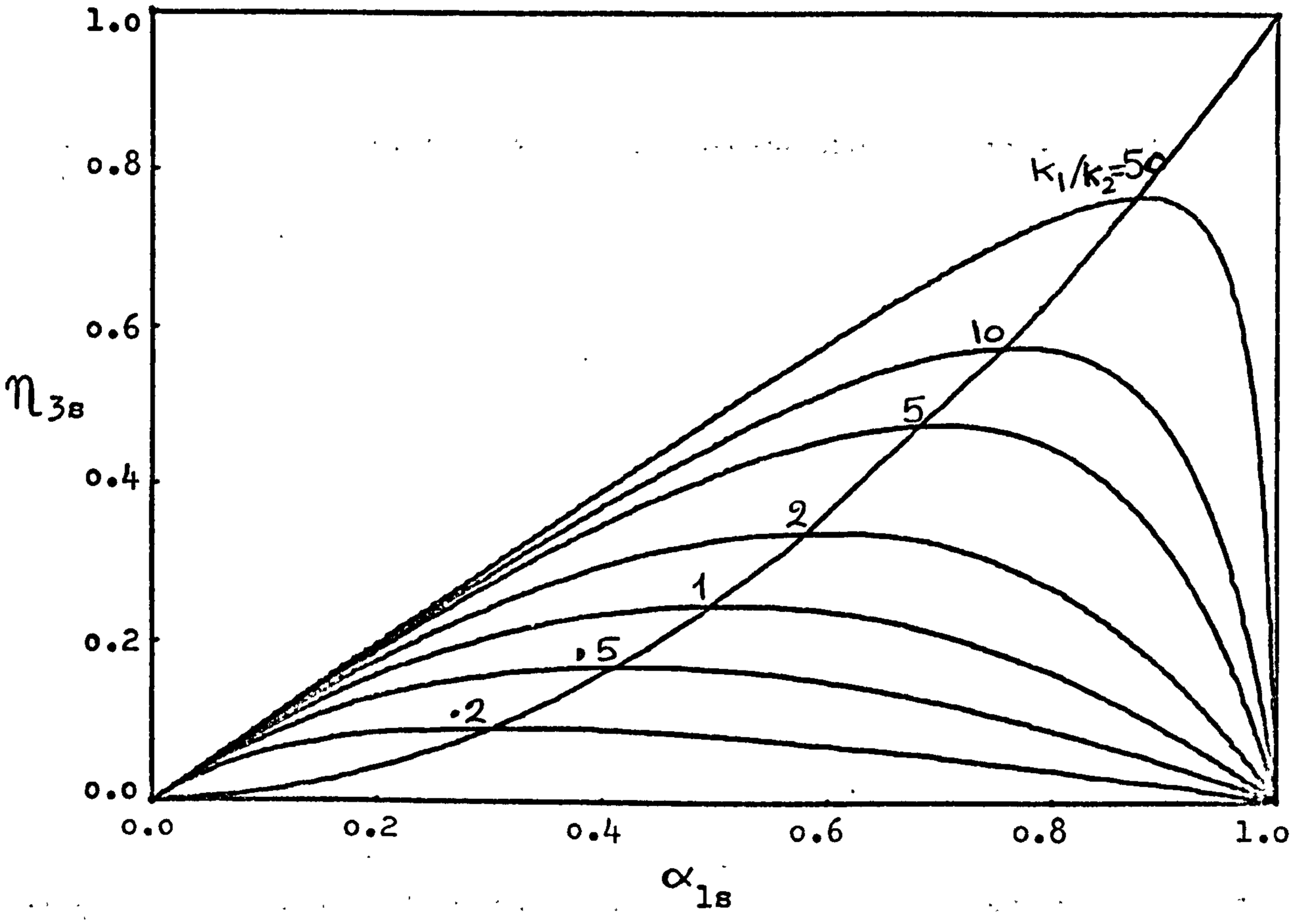


Fig. 2.1. Steady state yield of S_3 as a function of fractional conversion of S_1.

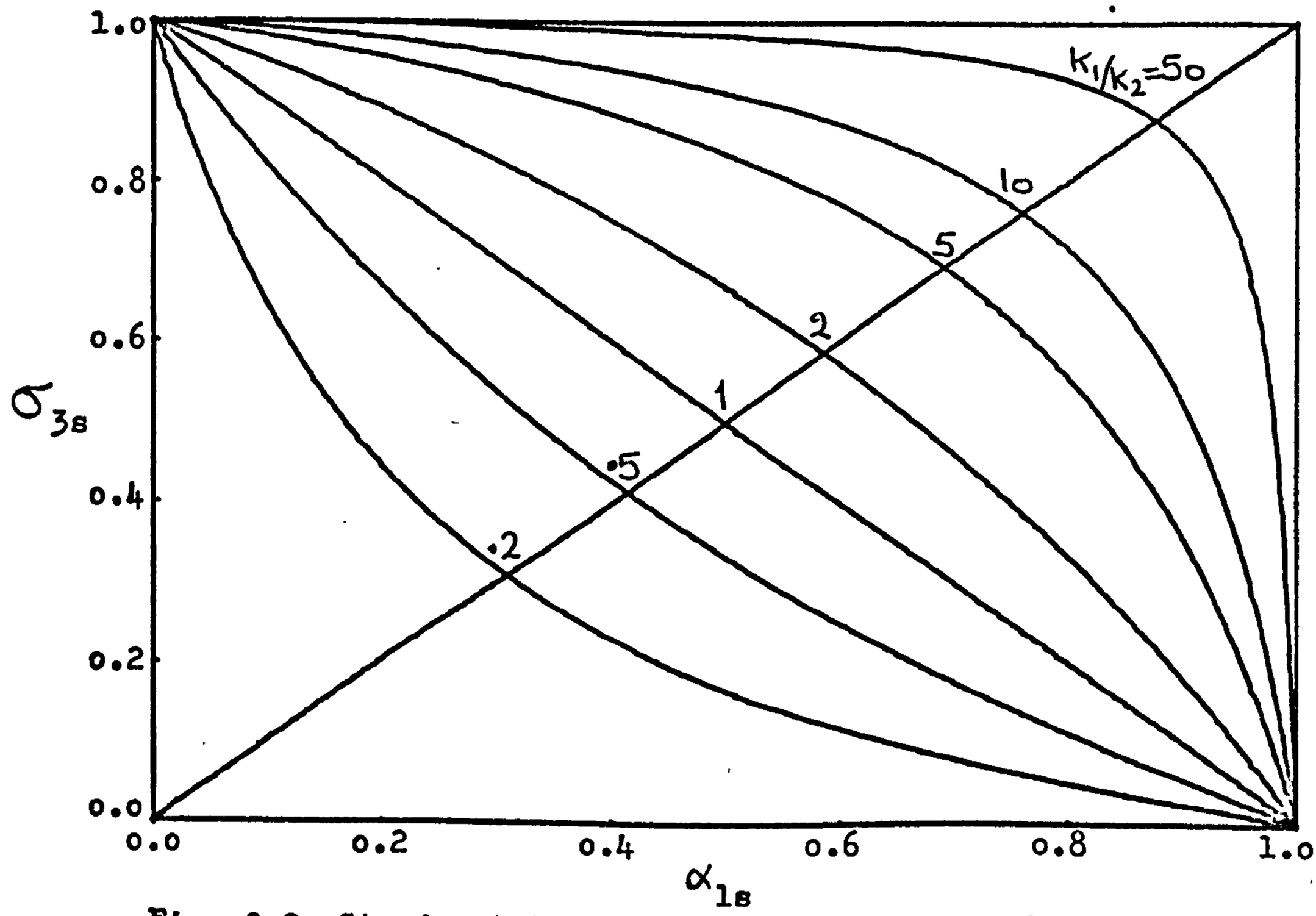


Fig. 2.2. Steady state selectivity of S_3 as a function of fractional conversion of S_1.

$$\dot{x}_1 = wu_1 - wx_1 - \alpha_{1s}x_1x_2$$

$$\dot{x}_2 = wu_2 - wx_2 - \alpha_{1s}x_1x_2 - \alpha_{2s}x_2x_3 \qquad 2.10$$

$$\dot{x}_3 = -wx_3 + \alpha_{1s}x_1x_2 - \alpha_{2s}x_2x_3$$

$$\dot{x}_4 = -wx_4 + \alpha_{2s}x_2x_3$$

where

$$x_i = A_i / A^*_{1fs}, i=1,2, \theta = tF^*_s/V$$

$$w = F/F^*_s, \alpha_{is} = K_i \exp(-\frac{Ei}{RT_s}) \frac{V}{F^*_s} A^*_{1fs}, i=1,2$$

The object is then to maximise the average yield of the desired product, S_3, defined by

$$\bar{\eta}_3 = \frac{\int_{\theta}^{\theta+\theta_p} wx_3 d\theta}{\int_{\theta}^{\theta+\theta_p} wu_1 d\theta}$$

The input concentration profiles are as shown in Figure 2.3 with the flow rate adjusted to

$$F = \frac{1}{\mu} F^*_s,$$

so that the same average amount of the reactants reach the reactor in all modes of operation. The case considered by Renken [46] is then obtained with μ = o.5. Furthermore, the total amount of reactant S_1 reaching the reactor in all modes is

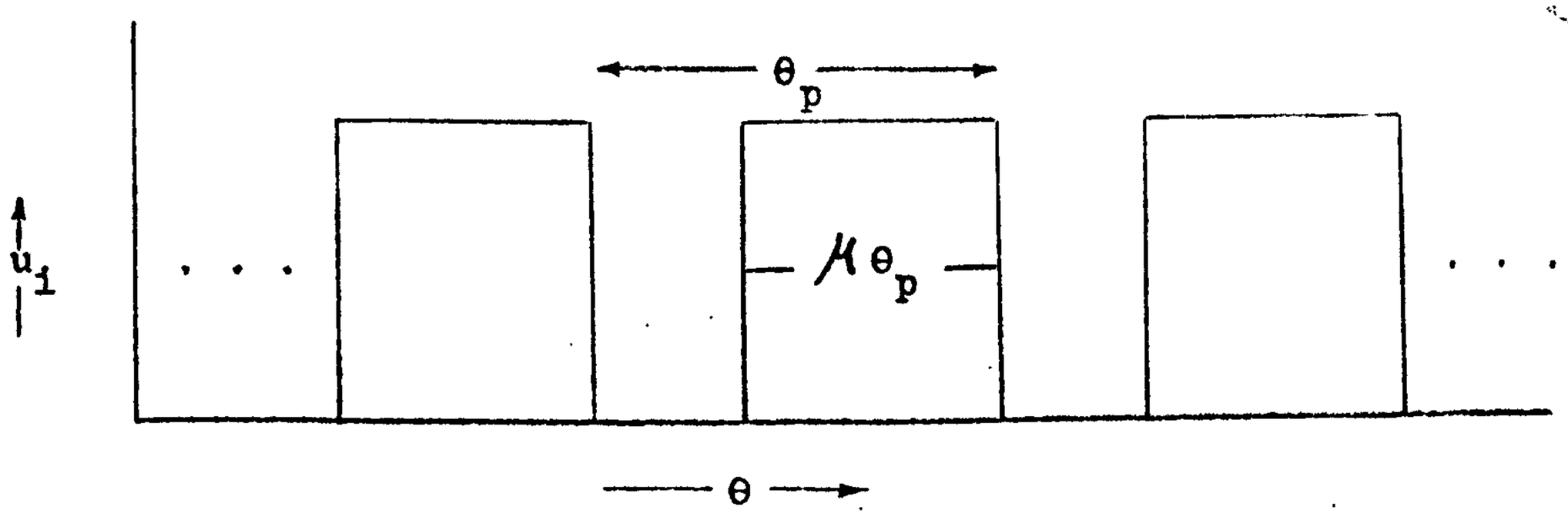


Fig. 2.3. Unsymmetrical square wave input profiles.

$$\int_{\theta}^{\theta+\theta_p} wu_1 d\theta = \theta_p,$$

and the objective may be written as

$$\bar{\eta}_3 = \frac{1}{\theta_p} \int_{\theta}^{\theta+\theta_p} wx_3 d\theta.$$

In practice, the above input profiles can be achieved by feeding the reactants to the reactor during each "on" fraction of a period with a flow rate, F, and feeding the diluent with the same flow rate during each "off" fraction.

The process was simulated on a digital computer using a variable step fourth order Runge-Kutta integration technique. The results obtained with μ = o.5 were identical to those of Renken [46]. However, as Figures 2.4 and 2.5 demonstrate further improvements are possible when unsymmetrical rather than symmetrical square waves are considered. A two dimensional search procedure for the best values of the input parameters, μ, Θ_p, for the case shown in Figure 2.4 yielded μ = o.65 and Θ_p = 2.75.

The simultaneous effect of periodic operation on the selectivity and the yield of the desired product is demonstrated in Figure 2.7. In this case at Θ_p = 1.5, the average yield, $\bar{\eta}_3$ = o.585, is just over one percent higher than the best achievable steady yield, η_s = o.577. The average selectivity, $\bar{\sigma}_3$ = o.882, however, exceeds the corresponding steady value, σ_s = o.760, by almost 16 percent. For the case shown in Fig. 2.6 at Θ_p = 1.25, the average yield, $\bar{\eta}_3$, is equal to the best achievable steady yield η_{3s}=o.25, the selectivity, $\bar{\sigma}_3$=o.630, on the other hand is some 26 percent higher than the corresponding steady value σ_s=o.50.

Thus for this idealised process, periodic operation is seen to be capable of improving both the quality and quantity of a desired product. The economic implication of these results for cases where the reactants are valuable and product separation difficult is apparent.

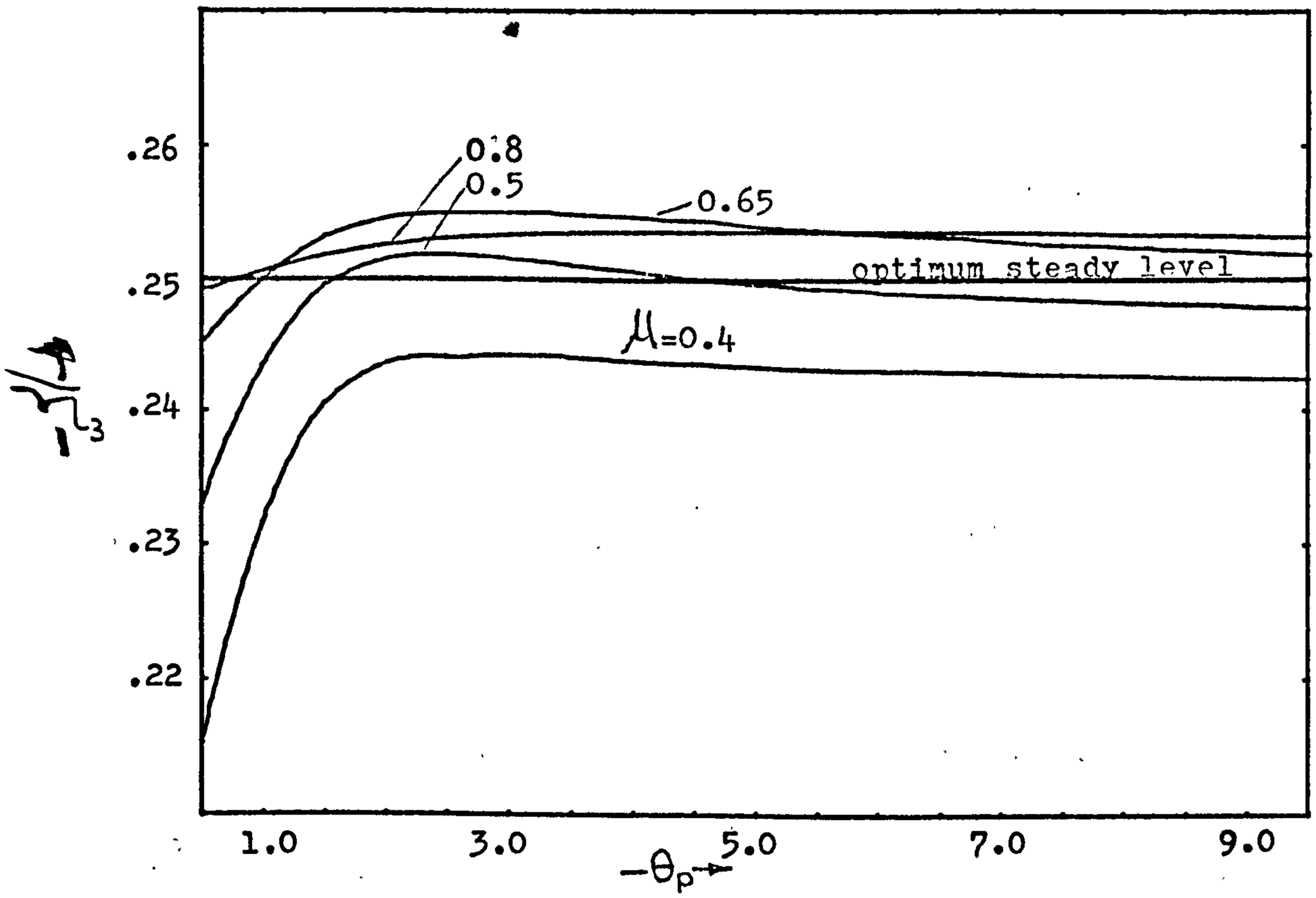


Fig.2.4. Average yield of S_3 as a function of the length of period for different values of μ.

(k_1/k_2= 1.0, A_{1fs}/A_{2fs}= 1.0, $k_1\tau_s A_{1fs}$= 4.0)

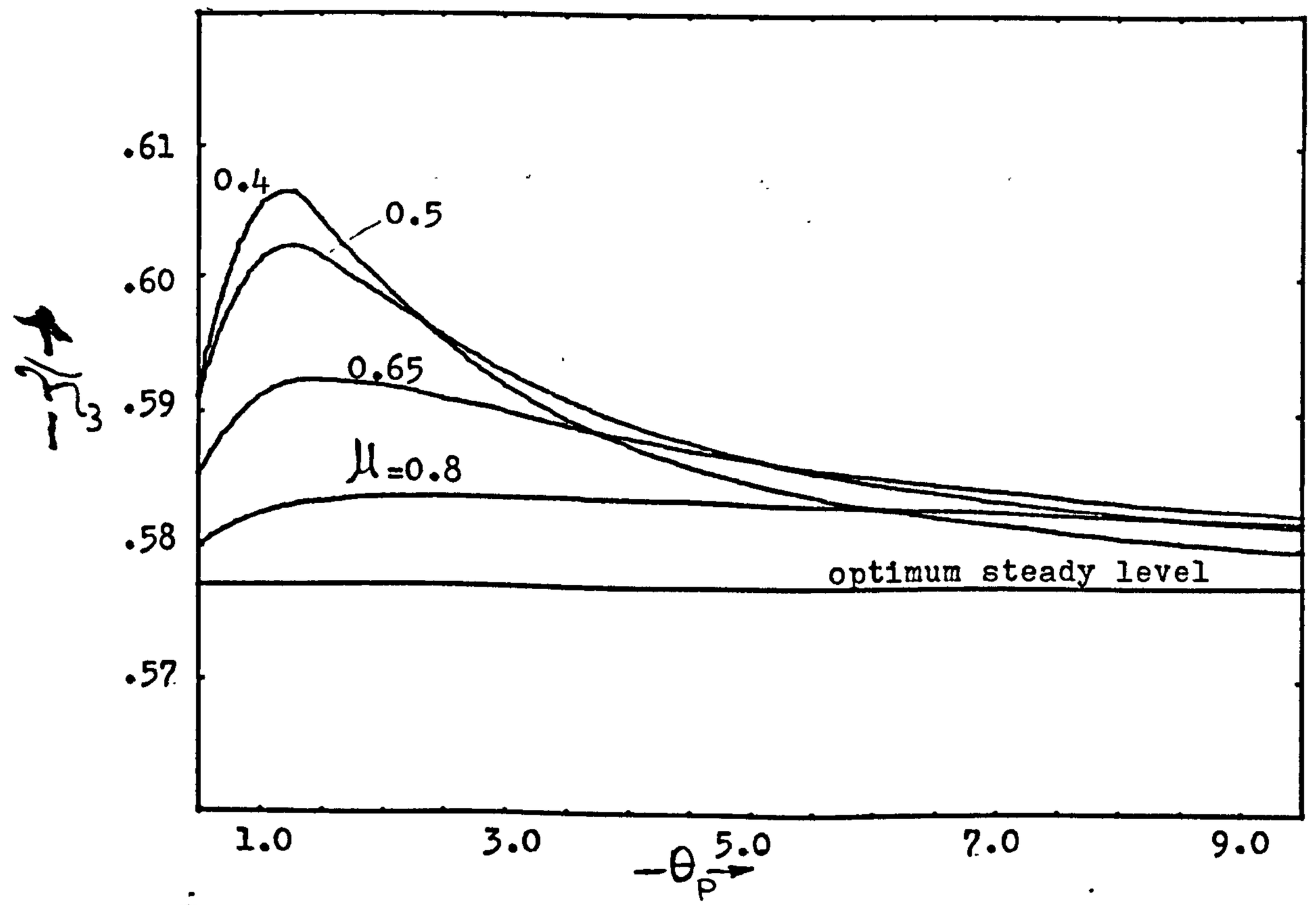


Fig.2.5. as figure 2.4 except

(k_1/k_2=10.0, $k_1\tau_s A_{1fs}$=54.785)

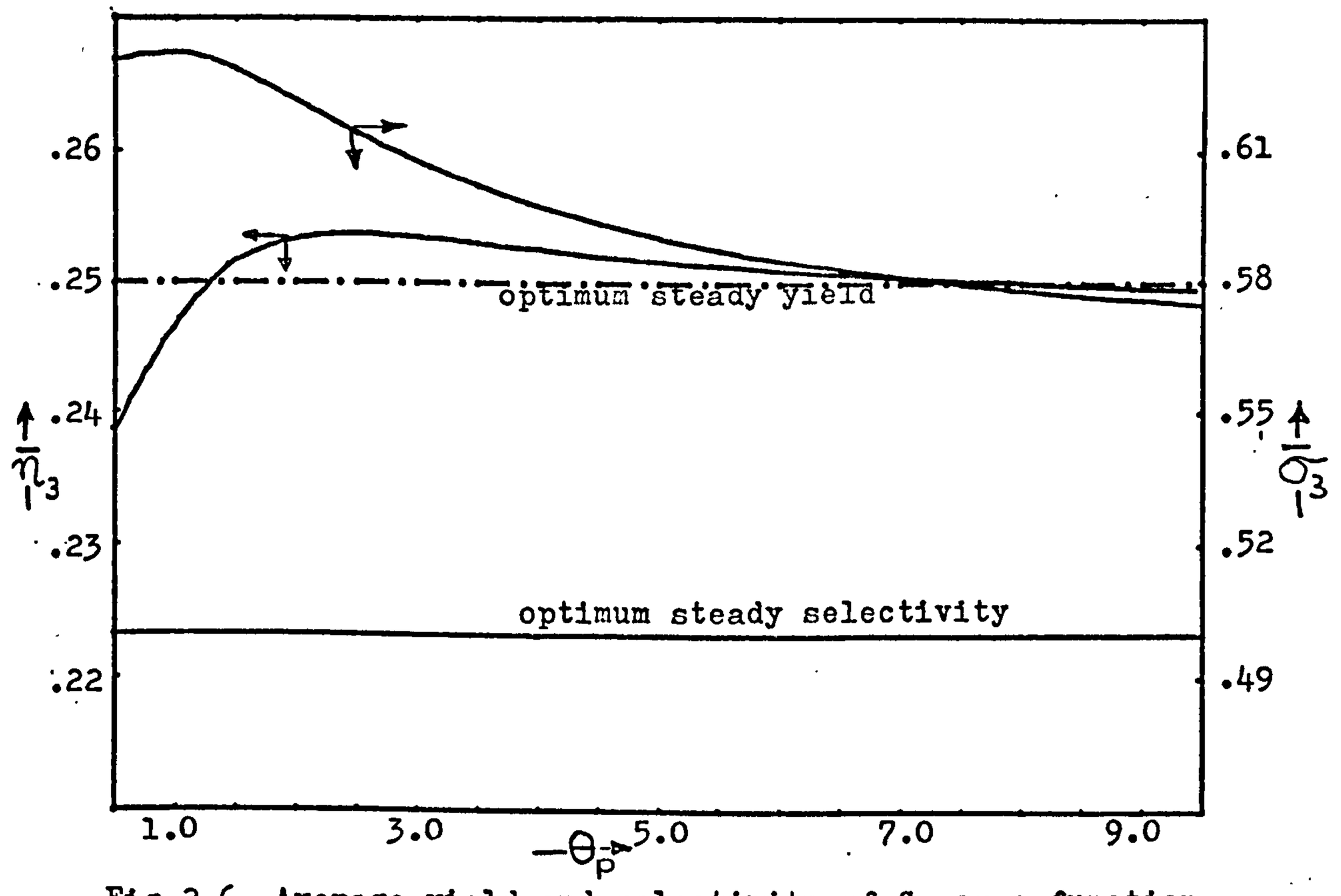


Fig.2.6. Average yield and selectivity of S_3 as a function of the length of period. (k_1/k_2= 1.0, μ= 0.55, A_{1fs}/A_{2fs}= 1.0, $k_1\tau_s A_{1fs}$= 4.0).

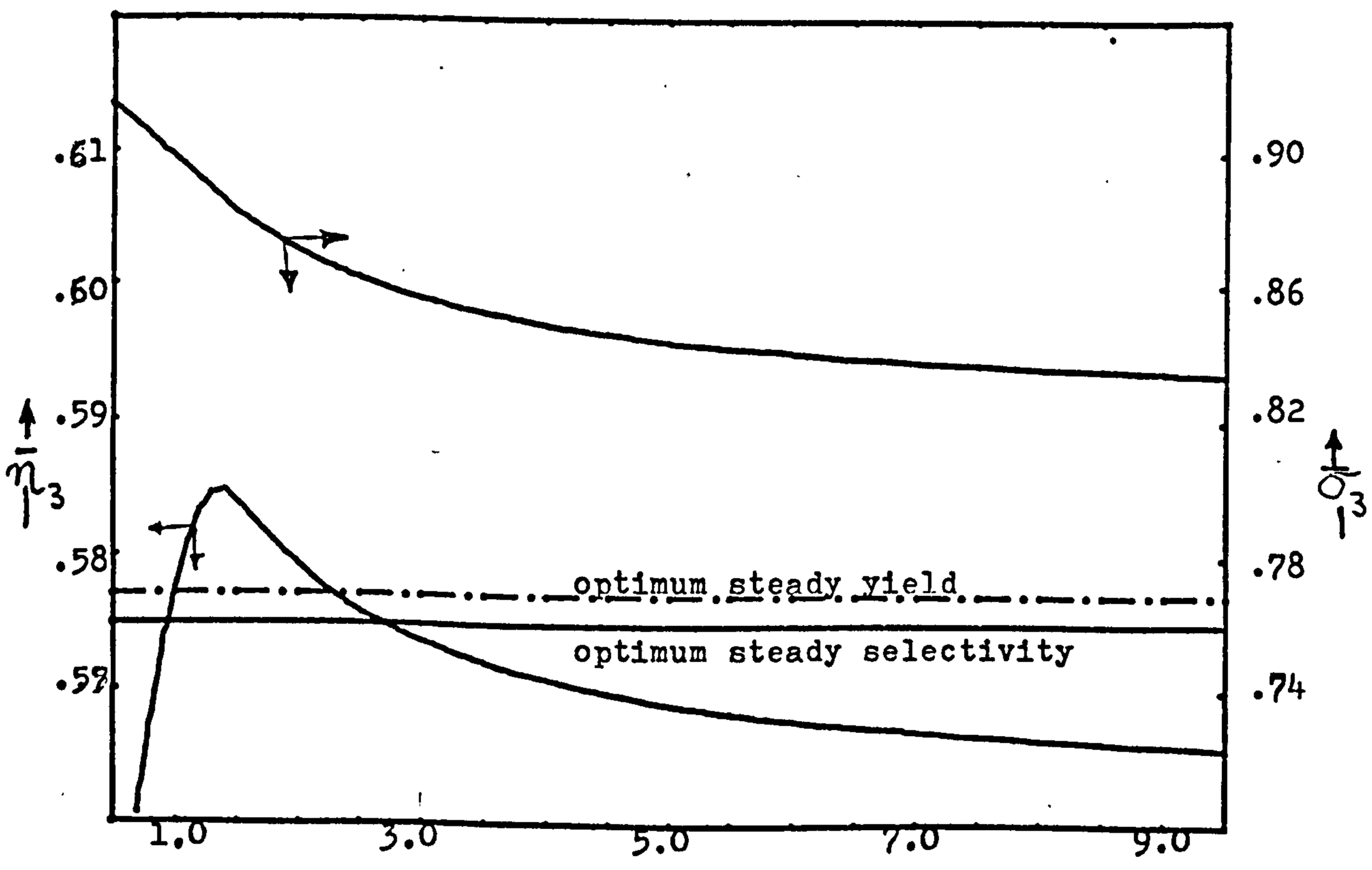


Fig.2.7. as figure 2.6 except

(k_1/k_2= 10.0, μ= 0.2, $k_1\tau_s A_{1fs}$=54.785).

2.3.2. The nonisothermal stirred tank reactor

The superior performance of a stirred tank reactor under periodic operation is due to the nonlinearity of the reaction rate expressions. In many isothermal reactors the rates are only mildly nonlinear and the improvements obtained are correspondingly small. In nonisothermal operations the inclusion of heat effects introduces an exponential nonlinearity into the rate equations and the magnitude of the expected profits of periodic operation are subsequently increased.

As an illustrative example the effect of symmetrical (μ = o.5) on-off variations of the feed composition to a nonisothermal stirred tank with reactions (2.9) was simulated. In this case the reactor is described by the following dimensionless system of equations:

$$\dot{x}_1 = wu_1 - wx_1 - \alpha_1 \exp(-1/x_5) x_1 x_2$$

$$\dot{x}_2 = wu_2 - wx_2 - \alpha_1 \exp(-1/x_5) x_1 x_2 - \alpha_2 \exp(-e/x_5) x_2 x_3$$

$$\dot{x}_3 = -wx_3 + \alpha_1 \exp(-1/x_5) x_1 x_2 - \alpha_2 \exp(-e/x_5) x_2 x_3$$

$$\dot{x}_4 = -wx_4 + \alpha_2 \exp(-1/x_5) x_2 x_3$$

$$\dot{x}_5 = w(x_{5f} - x_5) - h(x_5 - x_{5c}) + \beta_1 \exp(-1/x_5) x_1 x_2 + \beta_2 \exp(-1/x_5) x_2 x_3$$

where $x_i, i=1,\ldots,4, u_j, j=1,2$ and w are as in Eqs.(2.10) and the other dimensionless variables and groups are given by

$$x_5=RT/E_1, x_{5f}=RT_f/E_1, x_{5c}=RT_c/E_1, \quad h=U_a/F_s^*.C_p\rho,$$

$$\alpha_i=k_i\frac{V}{F_s^*}A_{1fs}^*, \beta_i=\alpha_i\frac{RA_{1fs}^*(-\Delta H_i)}{E_1C_p\rho}; i=1,2, e=E_2/E_1$$

The physical parameters used are given below Figure 2.8. and were chosen such that an isothermal operation at 500^oK corresponded to that shown in Figure 2.4. For small periods $\theta_p < 1.5$, the temperature within the tank does not rise much above the inlet temperature; a typical profile for the reactor is then as shown in Figure 2.8. For higher periods however, the temperature during the first half of the period rises to relatively high values before coming down as a result of rapid depletion of the reactants due to the very fast reactions which take place at these high temperatures. During the second half of the period the flow of reactants to the reactor is shut off and the temperature is reduced further. A typical profile for the reactor for this case is shown in Figure 2.9.

With the introduction of temperature the basis for comparison of steady and periodc operations becomes more difficult. For instance, with reaction schemes which are favoured by the highest possible temperature, the comparison of the periodic operation with a steady operation at the mean temperature is not strictly justified. Instead the results should be compared with a steady operation at the highest temperature during each period. For the case shown in Figure 2.9, the yield of the desired product, S_3, with periodic operation is o.2758, which is considerably higher than that of a steady operation at the mean temperature o.2344. It is however, significantly

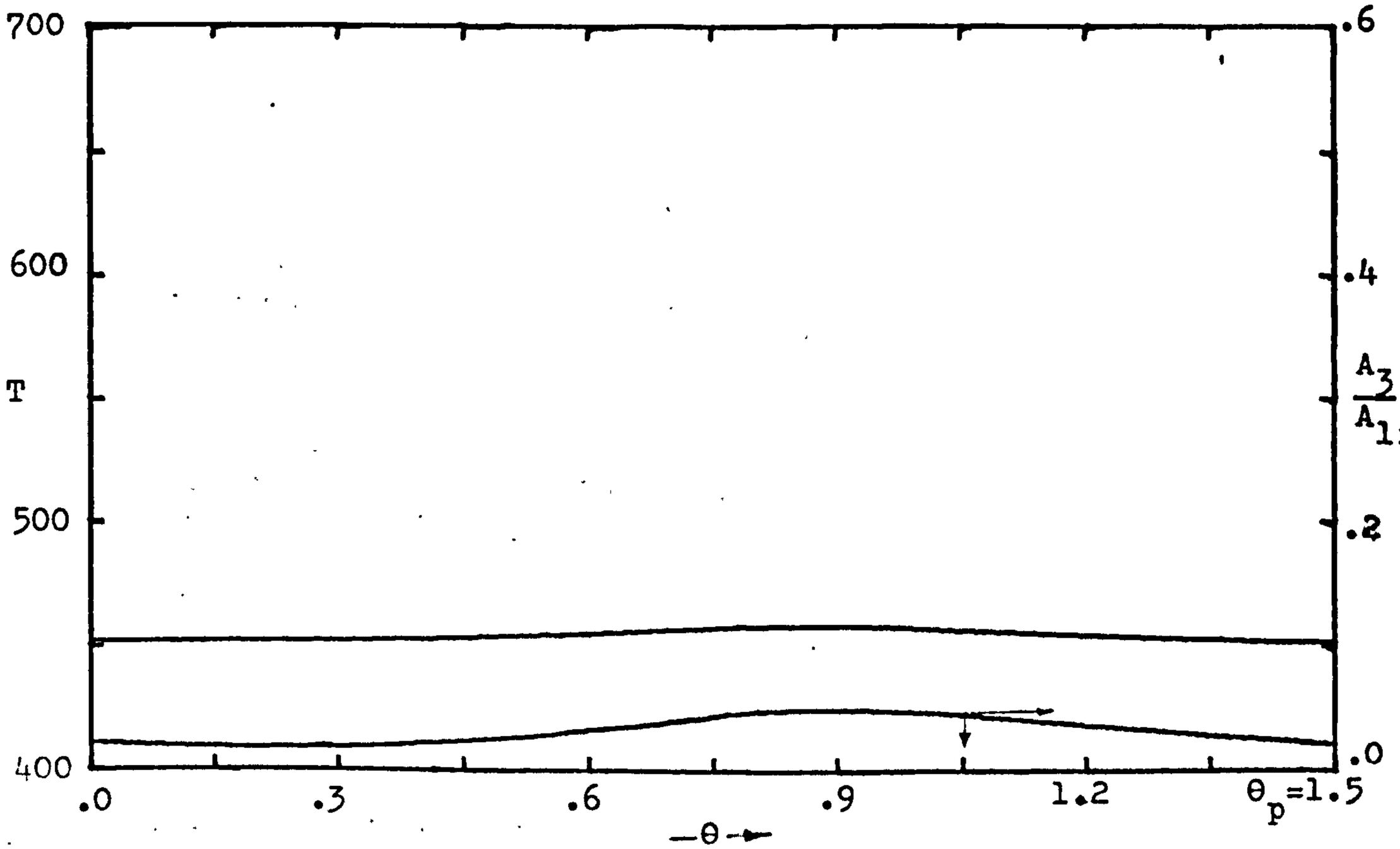


Fig.2.8. The temperature and composition of the desired product over one period.(α_1=4exp(25),α_2=4exp(20),e=0.8,h=2.75, $\beta_1=1.987\times10^{-2}$,$\beta_2=1.590\times10^{-2}$).

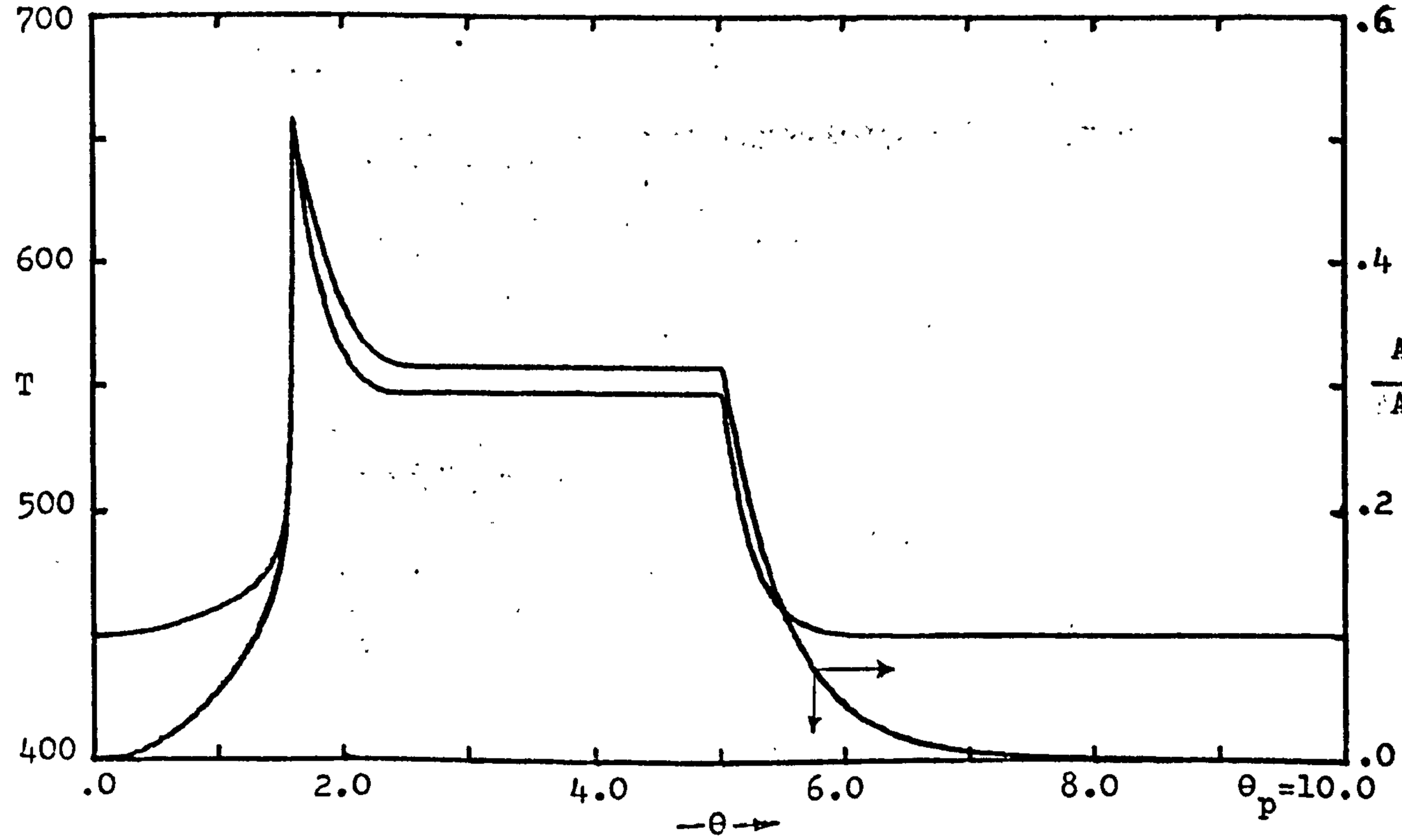


Fig.2.9. The temperature and composition of the desired product over one period.(parameters as in Fig.2.8.)

lower than the yield attainable with a steady operation at the highest temperature during the period, o.4537.

Another complication which could arise with nonisothermal operation is that of safety. Reaction run away may occur at high temperatures and there may well be material of construction limitations. So that, temperature overshoots of the type demonstrated in Figure 2.9 may have disastrous effects. In general, however, the proper choice of the periodic input can eliminate such problems; as is seen from Figure 2.8.

2.3.3. Long input sequences

So far we have arbitrarily chosen to consider on-off feed concentration profiles that are unimodal in the sense that only one switch occurs during each period. Clearly many alternatives are possible. For example, we could examine input profiles such as that given in Figure 2.10 which, as it happens, results in an improvement in the yield of the desired product over that obtained with the best symmetric square wave input considered by Renken [46].

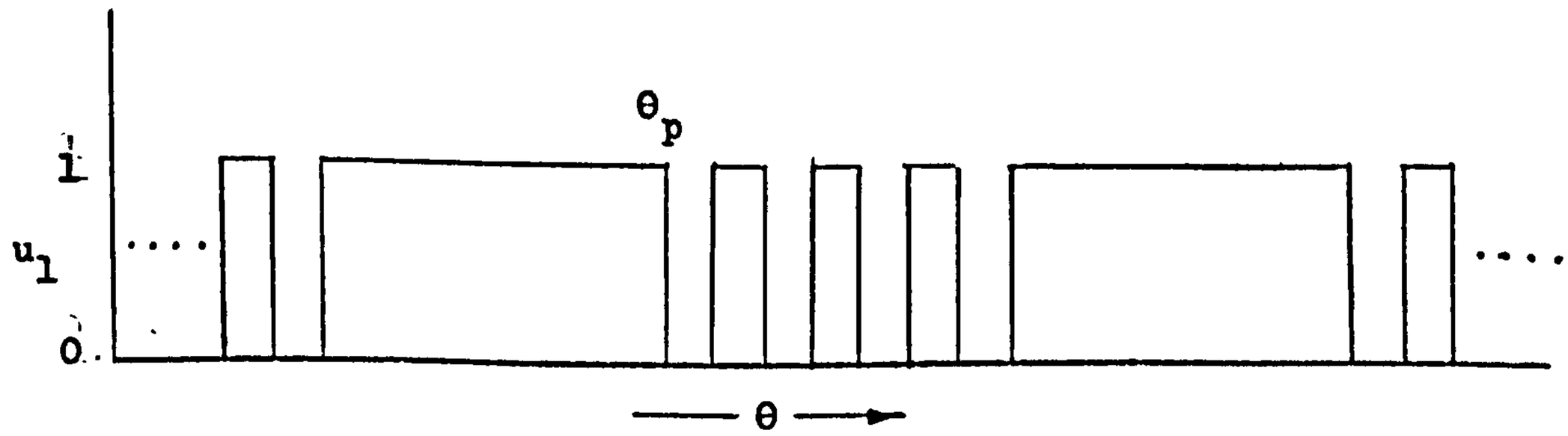


Fig.2.10. Periodic input concentration profiles of reactant S_1 (or S_2).

A further possibility would be to use a binary random input sequence which specifically excludes any repetative pattern. A commonly used and easily generated [65] sequence is the binary pseudo random one used in the dynamic testing of industrial processes. It is interesting to note that a process has effectively no memory of perturbations occuring more than a settling time before. So that, provided the random input sequence length is longer than a settling time, the average results obtained should be insensitive to the actual shape and length of the on-off random sequence used. This was indeed confirmed by the simulation results obtained for the isothermal stirred tank of section 2.3.1. The yield of the desired product, S_3, as a function of the minimum switching interval used went through a weak maximum which for the cases considered was inferior to steady operation.

Many other input profiles could be used; however considering the infinite number of unrelated alternatives, it is apparent that hit and miss methods are of little value in the determination of truly optimal concentration profiles. Furthermore, the basic assumption that the best profiles are on-off, switching between zero and the reference steady state level, has yet to be justified. It may also happen that the process inputs are best varied independently.

A rigorous approach is needed and the remaining chapters of this thesis are devoted to this. We shall first show that the mere formulation of the problem in accordance with optimal control theory provides considerable insight into the optimal strategy and answers some of the questions posed above.

We shall then proceed to examine and solve further problems that occur in the formulation and solution of problems of this type, finally arriving at a generalised computational algorithm for their efficient solution.

CHAPTER 3

UNSTEADY STATE OPERATION OF CHEMICAL REACTORS : A RIGOROUS APPROACH

In most applications a suboptimal scheme provides the practical answer to unsteady state processing. Such schemes can be found with relative computational ease and the procedure is in effect one of finding the extremum of a function of several constrained parameters. However, given a process and a cost criterion, there is only one way of finding what fraction of the potential benefits of unsteady processing is obtained with a suboptimal scheme and that is to determine the optimal mode of unsteady operation. The knowledge of the optimal scheme, as well as providing a yardstick for comparison of several suboptimal ones, is of great value to a designer who, in a real situation, will be seeking the most profitable and practical mode of operation while trying to compromise between several cost criteria.

The determination of optimal input profiles or control strategies is a variational problem which, unless the process is linear and the objective is of a simple form, must be solved numerically. In unsteady processing we are concerned with nonlinear processes and the numerical solution of the variational problem presents certain computational difficulties. The remainder of this thesis is concerned with the step by step identification of the difficulties involved and the development of a computationally efficient algorithm for finding optimal periodic modes of operation

Unfortunately much of the literature on variational methods has been presented in either a notationally confusing manner or at a mathematical level which obscures the simplicity of the basic

principles involved. A review paper by Douglas [66] provides a clear introduction to the variational problem and a comprehensive treatment of the subject may be found in the work of Athans and Falb [67]. The aim of this chapter is to introduce the basic principles involved and discuss, with the aid of an illustrative example, the difficulties which arise in the numerical solution of variational problems.

3.1. The basic theory

Consider a process described by

$$\frac{d\underline{x}}{dt} = \dot{\underline{x}} = \underline{f}(\underline{x},\underline{u},t), \underline{x}(t_o) \text{ given}, t_o \leqslant t \leqslant t_f, \qquad 3.1$$

where $\underline{x}(t)$, an n-vector function of state, is determined by the choice of $\underline{u}(t)$, an r- vector function of control, over the given time interval $t_o \leqslant t \leqslant t_f$.

Assume that the performance is measured through the scalar objective function

$$J = \psi(\underline{x}(t_f)) + \int_{t_o}^{t_f} f_o(\underline{x},\underline{u},t)dt, \qquad 3.2$$

where $f_o(\underline{x},\underline{u},t)$ is some measure of the instantaneous profit.

The problem is to find a control vector $\underline{u}^* = \underline{u}^*(t)$ which minimises the objective, J, while satisfying the system equations (3.1).

Following the classical procedure we adjoin the zero quantity

$$\underline{y}^T(t)\ (\dot{\underline{x}} - f(\underline{x},\underline{u},t)),$$

where $\underline{y}^T(t) = (y_1(t),\ldots,y_n(t))$ is a set of as yet undefined multiplier functions, to the objective, J, to yield:

$$J = \psi(\underline{x}(t_f)) + \int_{t_o}^{t_f} (f_o(\underline{x},\underline{u},t) + \underline{y}^T(\dot{\underline{x}} - \underline{f}(\underline{x},\underline{u},t))dt. \quad 3.3.$$

For convenience, we define a scalar function H (the Hamiltonian) as follows:

$$H = H\ (\underline{x}\ (t),\underline{y}(t),\underline{u}(t),t) = -f_o\ (\underline{x},\underline{u},t) + \underline{y}^T\ \underline{f}(\underline{x},\underline{u},t). \quad 3.4$$

Equation (3.3) can now be rewritten as

$$J = \psi(\underline{x}(t_f)) + \int_{t_o}^{t_f} (\underline{y}^T\dot{\underline{x}} - H)\ dt,$$

which on integrating the $\underline{y}^T\dot{\underline{x}}$ term by parts yields

$$J = \psi(\underline{x}(t_f)) + \underline{y}^T(t_f)\underline{x}(t_f) - \underline{y}^T(t_o)\underline{x}(t_o)$$

$$- \int_{t_o}^{t_f} (\dot{\underline{y}}^T\ \underline{x} + H)\ dt. \quad 3.5$$

Now for fixed times t_o and t_f, let us consider variations in J due to a small change in the control, $\delta\underline{u}(t)$, which in turn causes a small variation $\delta\underline{x}\ (t)$, in the state. Neglecting the second and higher order tems we have

$$\delta J = \left(\frac{\partial \psi}{\partial \underline{x}} + \underline{y}^T \right) \delta \underline{x}(t_f) - \underline{y}^T(t_o)\delta \underline{x}(t_o)$$

$$- \int_{t_o}^{t_f} \left((\dot{\underline{y}}^T + \frac{\partial H}{\partial \underline{x}}) \delta \underline{x}(t) + \frac{\partial H}{\partial \underline{u}} \delta \underline{u}(t) \right) dt. \qquad 3.6$$

It is tedious to determine the variation $\delta \underline{x}(t)$ caused by a given $\delta \underline{u}(t)$. This may be avoided by choosing

$$\dot{\underline{y}}^T = - \frac{\partial H}{\partial \underline{x}} = \frac{\partial f_o}{\partial \underline{x}} - \underline{y}^T \frac{\partial \underline{f}}{\partial \underline{x}} , \qquad 3.7\text{-a}$$

with the boundary conditions

$$\underline{y}^T(t_f) = -\left(\frac{\partial \psi}{\partial \underline{x}}\right)_{t_f} . \qquad 3.7\text{-b}$$

Substitution of Eqs. (3.7) into Eq. (3.6) results in the basic equation of calculus of variation

$$\delta J = -\underline{y}^T(t_o)\delta \underline{x}(t_o) - \int_{t_o}^{t_f} \frac{\partial H}{\partial \underline{u}} \delta \underline{u}(t) dt. \qquad 3.8$$

The physical significance of the multipliers, $\underline{y}(t)$, and the Hamiltonian, $H(\underline{x},\underline{y},\underline{u},t)$ are now clear: $\underline{y}(t_o)$ describes the effect of a change in the initial condition, $\underline{x}(t_o)$, on the objective while keeping the control constant, $\delta \underline{u}(t) = 0$. Furthermore, as the time t_o is arbitrary, $\underline{y}(t)$ represents the effect of a change in the state, $\delta \underline{x}(t)$, on the objective while keeping the control, $\underline{u}(t)$, constant. For this reason $\underline{y}(t)$ is often referred to as the vector of the influence functions. The functions

$$\frac{\partial H}{\partial u_j}(t),\ j=1,\ldots,r,$$

reflect the variation in J due to a unit impulse variation in the control variable, $u_j(t)$ at time t

$$\delta u_j(t) = \begin{cases} 1 \text{ at time } t \\ 0 \text{ at all other times} \end{cases}, \quad j=1,\ldots,r,$$

while keeping the initial conditions constant, $\delta \underline{x}(t_o)=o$. For this reason $\frac{\partial H}{\partial u_j}$'s are often referred to as the impulse response functions.

The system equations (3.1) and the multiplier or adjoint equations (3.7) can be written in a more compact form in terms of the Hamiltonian (see Eq.3.4)

$$\underline{\dot{x}}^T = \frac{\partial H}{\partial \underline{y}} = H_{\underline{y}},\ \underline{x}(t_o) \text{ given}$$

$$\underline{\dot{y}}^T = -\frac{\partial H}{\partial \underline{x}} = -H_{\underline{x}},\ \underline{y}^T(t_f) = -\left(\frac{\partial \psi}{\partial \underline{x}}\right)_{t_f} \qquad 3.9$$

3.1.1. The necessary conditions of optimality

We are now in a position to state the conditions which an optimal control must fulfil . Assuming the initial conditions do not vary (i.e. $\delta \underline{x}(t_o) = o$), the variation in the objective is given by

$$\delta J = -\int_{t_o}^{t_f} \frac{\partial H}{\partial \underline{u}}\, \delta \underline{u}(t)\, dt. \qquad 3.10$$

Now, if the controls are unrestricted, $\delta\underline{u}(t)$ can have any arbitrary small value. Let us take

$$\delta\underline{u}(t) = \varepsilon\left(\frac{\partial H}{\partial \underline{u}}\right)^{T} \qquad 3.11$$

Where ε is a positive constant. Equation (3.10) then becomes,

$$\delta J = -\varepsilon \int_{t_o}^{t_f} \left\| \frac{\partial H}{\partial \underline{u}} \right\|^2 dt$$

which is always non-positive. The objective can therefore be reduced in all cases except when,

$$\frac{\partial H}{\partial \underline{u}}(t) = o, \text{ for all } t,\ t_o \leqslant t \leqslant t_f, \qquad 3.12$$

which provides r necessary conditions which the r control variables must satisfy.

In practice, the control variables cannot take on arbitrary values and are restricted to certain physically realisable limits. Let us assume that the control constraints are given by

$$u_j^{min} \leqslant \dot{u}_j(t) \leqslant u_j^{max}, \text{ for all } t,\ t_o \leq t \leq t_f, j = 1,\ldots,r.$$

In this case, the variations, $\delta\underline{u}(t)$, are no longer arbitrary and must be such that the constraints are not violated. It is clear that along those portions of the control history which lie entirely within the constraints, any sufficiently small control perturbation is admissible

and the necessary conditions are given by Eq. (3.12) as before. It remains to examine those portions of the control history which lie entirely on the boundary.

A brief examination of Eq. (3.10), which is valid for all cases, reveals that we must have

$$\frac{\partial H}{\partial \underline{u}} \delta \underline{u}(t) \leqslant o, \tag{3.13}$$

for all admissible $\delta \underline{u}(t)$. Another way of stating Eq. (3.13) is that improvements should only be possible through the violation of the boundaries.

Now consider the time derivative of the Hamiltonian; the application of the chain rule yields

$$\frac{d}{dt} H(t) = \dot{H} = \frac{\partial H}{\partial t} + \frac{\partial H}{\partial \underline{x}} \dot{\underline{x}} + \frac{\partial H}{\partial \underline{y}} \dot{\underline{y}} + \frac{\partial H}{\partial \underline{u}} \dot{\underline{u}}$$

which using Eqs. (3.9) reduces to

$$\dot{H}(t) = \frac{\partial H}{\partial t} + \frac{\partial H}{\partial \underline{u}} \cdot \dot{\underline{u}} \tag{3.14}$$

Then, if the function f_o and the vector function $\underline{f}$ are not explicit functions of time $\frac{\partial H}{\partial t} = o$ by definition (see Eq. 3.4). Furthermore, over these portions of the optimal control which lie inside the boundary $\frac{\partial H}{\partial \underline{u}}(t) = o$, and for those portions which lie entirely on the boundary

$\dot{\underline{u}}(t) = 0$. So that in any event, $\frac{dH}{dt}(t) = 0$, from which we obtain

$$H(\underline{x},\underline{y},\underline{u}) = \text{a constant}, \qquad 3.15$$

over the entire period $t_o \leq t \leq t_f$.

Actually, a much stronger statement of the optimality principle is possible: this is that the Hamiltonian, $H(\underline{x},\underline{y},\underline{u},t)$, viewed as a function of the control $\underline{u}$, alone must be maximum at each time t. This compact statement is due to Pontryagin [68] and is known as the Maximum Principle. The mathematical justification of the Maximum Principle is extremely complicated and falls outside the scope of this thesis. A rigorous proof of this principle, taking into account strong variations and restrictions on the terminal state, is given by Pontryagin and his co-workers [68]. A heuristic proof is supplied by Athans and Falb [67] and Douglas [66] has given a lucid introduction to the basic principles involved. We shall first give a general statement of the principle and then examine its application to a specific problem in unsteady state processes.

3.1.2. The statement of Maximum Principle

Consider a process described by Eqs. (3.1) and an objective given by Eq. (3.2) and assume that $\underline{f}(\underline{x},\underline{u},t)$ and $f_o(\underline{x},\underline{u},t)$ are continuously differentiable functions of their arguments and $\underline{u}(t)$ is restricted to a certain physically realisable set of values.

Then if an optimal control, $\underline{u}^*(t)$, exists it must be such that the adjoint variables y_o and $\underline{y}(t)$ defined through

$$\dot{y}_o = o,$$
$$\dot{\underline{y}} = -\left(\frac{\partial f_o}{\partial \underline{x}}\right)^T y_o - \left(\frac{\partial \underline{f}}{\partial \underline{x}}\right)^T \underline{y}, \underline{y}(t_f) = -\left(\frac{\partial \psi(\underline{x})}{\partial \underline{x}}\right)^T_{t_f}, \qquad 3.16$$

and the Hamiltonian, H, given by

$$H(\underline{x},\underline{y},y_o,\underline{u},t) = f_o(\underline{x},\underline{u},t)y_o + \underline{y}^T\underline{f}(\underline{x},\underline{u},t), \qquad 3.17$$

satisfy the following conditions:

I. y_o^* is a non-positive constant;

II. The vector $(y_o^*, \underline{y}^{*T}(t)) \neq o$;

III. The Hamiltonian viewed as a function of the control, $\underline{u}$, alone attains its largest possible value at the point $\underline{u}=\underline{u}^*(t)$ at all times $t, t_o \leqslant t \leqslant t_f$.

It also turns out that if $f_o(\underline{x},\underline{u},t)$ and $\underline{f}(\underline{x},\underline{u},t)$ are not explicit functions of time, the Hamiltonian assumes a constant value along the optimal trajectory. Furthermore, the adjoint system and the Hamiltonian (see Eqs. 3.16 and 3.17) are linear in y_o^*, so that if $y_o^* \neq o$ one may without loss of generality set it to -1:

$$y_o^* = -1.$$

Often the mere formulation of the problem in terms of the Maximum Principle yields valuable information about the structure of the optimal control strategies. As an example consider the problem of finding the optimal input concentration profiles for periodic operation of an isothermal stirred tank reactor discussed in chapter 2. For this problem, the state system is given by Eqs. (2.10) which are repeated for convenience:

$$\begin{aligned}
\dot{x}_1 &= wu_1 - wx_1 - (k_1\tau_s A_{1fs})x_1x_2, \\
\dot{x}_2 &= wu_2 - wx_2 - (k_1\tau_s A_{1fs})x_1x_2 - (k_2\tau_s A1fs)x_2x_3, \\
\dot{x}_3 &= -wx_3 + (k_1\tau_s A_{1fs})x_1x_2 - (k_2\tau_s A_{1fs})x_2x_3, \\
\dot{x}_4 &= -wx_4 + (k_2\tau_s A_{1fs})x_2x_3.
\end{aligned} \tag{3.18}$$

The object is to maximise the yield of the desired product or minimise the integral functional

$$J = -\frac{1}{\theta_p}\int_0^{\theta_p} wx_3 d\theta \tag{3.19}$$

subject to the conditions that the inlet concentrations do not exceed the corresponding steady level,

$$o \leqslant u_j(\theta) \leqslant 1, \text{ for all } \theta, \; o \leqslant \theta \leqslant \theta_p, j=1,2, \tag{3.20}$$

and equal average amounts of the reactants reach the reactor in all modes of operation:

$$\int_0^{\theta_p} wu_j(\theta)d\theta = \theta_p,\ j = 1,2. \qquad 3.21$$

The introduction of two new state variables

$$\dot{x}_5 = wu_1, x_5(o) = o, x_5(\theta_p) = \theta_p,$$
$$\dot{x}_6 = wu_2, x_6(o) = o, x_6(\theta_p) = \theta_p, \qquad 3.22$$

reduces the problem to the form considered above, and the Hamiltonian can be written:

$$H = w(y_1+y_5)u_1+w(y_2+y_6)u_2+$$
$$-w(\frac{1}{\theta_p}y_o x_3+y_1x_1+y_2x_2+y_3x_3+y_4x_4)$$
$$-(k_1\tau_s A_{1fs})x_1x_2(y_1+y_2-y_3)$$
$$-(k_2\ _s A_{1fs})x_1x_2(y_2+y_3-y_4)$$

The control variables appear only in the first two terms of this expression and it is obvious that, for given $x_i(\theta)$ and $y_i(\theta)$, H achieves its maximum value when these variables, u_1 and u_2, switch between their extreme values, that is between zero and one, according to the sign of the functions (y_1+y_5) and (y_2+y_6) respectively.

The optimal input profiles are thus seen to be on-off, switching between zero and the maximum (steady state) values. There is, however, no reason to assume that the optimal on-off controls should have the unimodal periodic form considered in the earlier empirical approach, or that they should switch in phase. The truly optimal strategy must be obtained by solving the problem posed above; this is of a delicate two-point boundary-value nature and its numerical solution, as we shall see later, is beset with difficulties.

3.1.3. Limiting periodic operations: relaxed steady state analysis

The application of the Maximum Principle to an optimal steady process provides an easy test for prediction of the possible improvements through dynamic operation. Such tests are of great value and whenever possible should precede a detailed study. Another technique which provides an easy test is the relaxed steady state analysis which involves the study of very high frequency periodic operations. To this end, limiting periodic operations merit a closer look.

This problem was first examined by Horn and Lin [47] who showed that rapid switching of temperature inside a stirred tank reactor results in significant improvements over the optimal steady operation. Since then this type of operation has been shown to be capable of improving catalytic selectivity[48,61,62].

The mathematical analysis of this problem is due to Warga [71] and essentially similar results are obtained by Rinaldi [72]

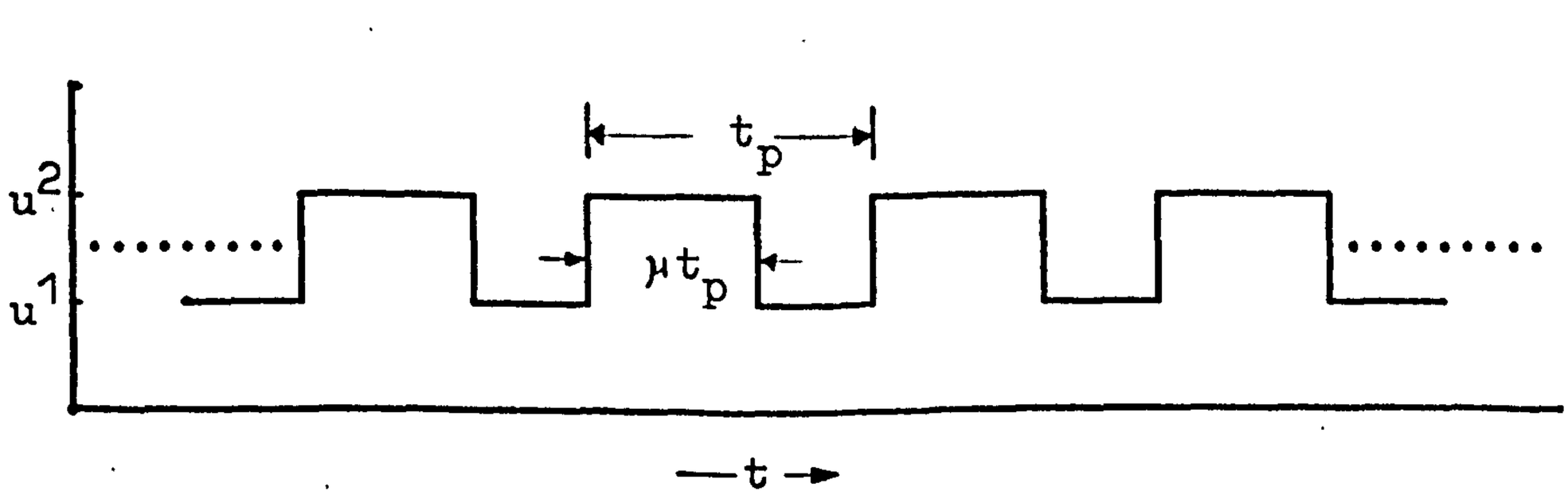
who has applied the Maximum Principle to the determination of optimal high frequency operations. The results indicate that the optimal input profile is piecewise constant and switches between admissible values during each period. An upper bound on the number of switches has also been derived [72]. Although the detailed mathematics is quite involved, the basic ideas are quite simple and are best understood in the following manner.

Consider a process with a single input, u, whose dynamics are governed by

$$\dot{\underline{x}} = \underline{f}(\underline{x}, u),$$

and let the input be as shown below, so that during each period

$$u = \begin{cases} u^2, & \text{for } o \leqslant t < \mu t_p, \\ u^1, & \text{for } \mu t_p < t \leqslant t_p. \end{cases}$$



Binary on-off input profile.

The state at the end of each period, $\underline{x}(t+t_p)$ is then related to the state at its beginning, $\underline{x}(t)$, through

$$\underline{x}\ (t+t_p) = \underline{x}(t) + \int_t^{t+\mu t_p} \underline{f}(\underline{x},u^1)dt + \int_{t+\mu t_p}^{t+t_p} \underline{f}(\underline{x},u^2)dt. \qquad 3.24$$

Now if the process is truly periodic, $\underline{x}(t+t_p) = \underline{x}(t)$, and Eq. (3.24) reduces to

$$0 = \int_t^{t+\mu t_p} \underline{f}(\underline{x},u^1)dt + \int_{t+\mu t_p}^{t+t_p} \underline{f}(\underline{x},u^2)dt. \qquad 3.25$$

If the period, t_p, is sufficiently long, with the exception of the transition intervals corresponding to switches in the control, the process is either in a steady operation corresponding to u^1 or one corresponding to u^2. In the limiting case (as $t_p \rightarrow \infty$) the contributions from the transition intervals are small, in comparison with those from the infinitely long steady operations, and can be ignored. As a result , a slow frequency operation is analogous to the mixing of steady states and as such cannot be superior to the optimal steady operation. A detailed analysis of this limiting case is given by Locatelli and Rinaldi [73].

If on the other hand the period, t_p, is made progressively smaller, the process is less and less able to follow the input variations and in the limiting case ($t_p \rightarrow o$) assumes a practically constant or relaxed steady state

$$\lim_{t_p \to 0} \underline{x}(t) \longrightarrow \hat{\underline{x}} \quad \text{a constant state.}$$

Equation (3.25) can then be integrated and becomes

$$o = \mu \underline{f}(\hat{\underline{x}}, u^1) + (1 - \mu)\underline{f}(\hat{\underline{x}}, u^2). \qquad 3.26$$

It is interesting to note that in this limiting case only the shape of the input is important and the actual magnitude of the period, t_p, is of no consequence, provided it is sufficiently small. Naturally, if the vector function $\underline{f}(\hat{\underline{x}},u)$ is linear in the control variable the relaxed steady state coincides with an ordinary steady state obtained with $u = \mu u^1 + (1 - \mu)u^2$, and as such cannot be better than an optimal steady state.

However, if the vector function $\underline{f}(\underline{x},u)$ is nonlinear with respect to the control variable the relaxed steady state may differ from an ordinary one. In fact it may happen that there is an optimum μ^* and a corresponding relaxed steady state $\hat{\underline{x}}^*$ which cannot be obtained with any single valued steady control; $\hat{\underline{x}}$ may therefore be superior to the optimal steady state. This line of reasoning is straightforwardly extended to any other piecewise constant periodic input and more than one control variable. The actual solution of the problem can be carried out via mathematical programming as discussed in detail by Bailey and Horn [61] and Rinaldi [72].

In conclusion it should be pointed out that high frequency operations prove superior in idealised cases when the process offers no resistance to the input variations. In general, when the process has damping effects, improvements are obtained

not with a very high but with a finite frequency. These ideas are discussed at length in the literature [48,61,62]. Furthermore, although it is true that in certain cases [74] relaxed steady state indicates improvements when the Maximum Principle does not, the latter theorem is more useful for the synthesis of optimal periodic controls.

The first chemical engineering application of the Maximum Principle appears to be due to Katz [76]. Since then it has been applied to the start up problem for stirred tank, tubular and batch reactors [77,78,79]. The application of the Maximum Principle to the determination of the optimal temperature profile in a tubular reactor is considered by Edward and Jackson [80], and its application to recycle reactors has been examined in several publications [81,82,83]. The determination of optimal periodic input profiles for unsteady operation of chemical reactors was first discussed by Horn and Lin [47] and we shall now consider a specific application of the Maximum Principle to this problem.

3.2. The application of the Maximum Principle to the determination of optimal unsteady operations.

By means of digital simulation Renken [46] demonstrated that the switching of reactant concentration to a stirred tank reactor in unimodal-periodic manner between a reference steady level and zero can result in improvements over the optimal steady performance for isothermal consecutive-competing reaction shcemes. In section 3.1.2. it was shown that the optimum mode of operation for this system is indeed likely to consist of switches between the minimum and maximum permissible reactant concentration levels, but that no reason exists for confining the search to switches of a unimodal-periodic nature beyond the obvious practical one that, in the face of an infinity of unrelated alternatives, some restrictions that enable a systematic search to be made become essential.

Our purpose here is to lift these constraints and, by a formal application of optimal control theory, arrive at operating policies that satisfy the necessary conditions of optimality. To simplify matters somewhat a modified objective function that relates to both reactor selectivity and production rate is employed. The problem is then solved for cases where the reactant concentrations are constrained to vary together, and also where this restriction is relaxed and the inputs are allowed to vary independently. The numerical results obtained as well as confirming the empirical approach previously employed [46,75], demonstrate some of the difficulties associated with

the numerical solution of optimal control problems of the type considered here.

3.2.1. The reaction scheme

We consider the same reaction system as that of references [46] and [75] namely

$$S_1 + S_2 \xrightarrow{r_1} S_3 \ , \ r_1 = k_1 A_1 A_2$$

$$S_2 + S_3 \xrightarrow{r_2} S_4 \ , \ r_2 = k_2 A_2 A_3$$

carried out isothermally in a continuous flow stirred tank reactor with S_3 taken as the desired product.

This process is described by the following dimensionless equations

$$\begin{aligned}
\dot{x}_1 &= wu_1 - wx_1 - \alpha_1 x_1 x_2, \ x_1(o) = o \\
\dot{x}_2 &= wu_2 - wx_2 - \alpha_1 x_1 x_2 - \alpha_2 x_2 x_3, \ x_2(o) = o \\
\dot{x}_3 &= -wx_3 + \alpha_1 x_1 x_2 - \alpha_2 x_2 x_3, \ x_3(o) = o \\
\dot{x}_4 &= -wx_4 + \alpha_2 x_2 x_3, \ x_4(o) = o
\end{aligned} \qquad 3.27$$

where the following normalisations have been made: all concentrations are measured with reference to the optimal steady input concentration of reactant S_1, A_{1fs}; flow rate is referred to the optimum steady state flow rate, F_s, and

time to the optimum steady mean residence time, τ_s. The dimensionless variables are then as defined in Eqs. (2.10).

3.2.2. The objective function

We adopt as our objective the minimisation of the integral functional J,

$$J = -\frac{1}{\theta_f}\int_o^{\theta_f} w(x_3 - x_4)d\theta, \qquad 3.28$$

which represents the difference between the production rates of the desired, S_3, and undesired, S_4, products over a sufficiently long time interval θ_f. This is a rather convenient expression of reactor performance as it depends on both the production rate of S_3 and the selectivity without involving the incorporation of a control effort restriction and ratio-integral objectives which, as we shall see later, are called for in a strict comparison between steady and dynamic modes of operation.

The objective (Eq. 3.28) may be incorporated into the state equations (3.27) by the introduction of an additional state variable defined by

$$\dot{x}_o = -w(x_3 - x_4),\ x_o(o) = o, \qquad 3.29$$

so that $x_o(\theta_f) = J$, and the problem is readily formulated for solution in accordance with the Maximum Principle as outlined above and fully described in [68].

3.2.3. The adjoint system and the Hamiltonian

The adjoint equations obtained from

$$\dot{\underline{y}}^T = -\frac{\partial f_o(\underline{x},\underline{u})}{\partial \underline{x}} - \underline{y}^T \frac{\partial \underline{f}(\underline{x},\underline{u})}{\partial \underline{x}} \; ; \; \underline{y}^T(\theta_f) = -\left(\frac{\partial \psi}{\partial \underline{x}}\right)_{\theta_f}$$

are thus given by

$$\begin{aligned}
\dot{y}_o &= o, \; y_o \leqslant o, \\
\dot{y}_1 &= w y_1 + \alpha_1 x_2 (y_1 + y_2 - y_3) \; , \; y_1(\theta_f) = o, \\
\dot{y}_2 &= w y_2 + \alpha_1 x_1 (y_1 + y_2 - y_3) + \alpha_2 x_3 (y_2 + y_3 - y_4), y_2(\theta_f) = o, \qquad 3.30 \\
\dot{y}_3 &= w(y_3 + y_o) + \alpha_2 x_2 (y_2 + y_3 - y_4), \; y_3(\theta_f) = o, \\
\dot{y}_4 &= w(y_4 - y_o), \; y_4(\theta_f) = 0,
\end{aligned}$$

and the Hamiltonian is given by

$$\begin{aligned}
H(\underline{x},\underline{y},y_o,\underline{u}) = \; & w y_1 u_1 + w y_2 u_2 \\
& - w(y_o x_4 - y_o x_3 + y_1 x_1 + y_2 x_2 + y_3 x_3 + y_4 x_4) \\
& - \alpha_1 x_1 x_2 (y_1 + y_2 - y_3) \\
& - \alpha_2 x_2 x_3 (y_2 + y_3 - y_4) \qquad 3.31
\end{aligned}$$

In this case we may take $y_o = -1$ without loss of generality. Then, the optimality condition is that the variables u_1 and u_2 are chosen so that at all times H takes on its maximum possible value. That is to say they take on their extreme values according to the signs of y_1 and y_2 respectively.

3.2.4. Method of solution

The application of variational methods invariably yields a two-point boundary value problem whose numerical solution proves the major obstacle in the determination of optimal control strategies.

The differential system (3.7) which defines the multiplier functions, $y_i(t)$, is by definition adjoint to the perturbation equations obtained by linearisation of the state system (3.1):

$$\delta\dot{\underline{x}} = \frac{\partial \underline{f}(\underline{x},\underline{u},t)}{\partial \underline{x}}\,\delta\underline{x} + \frac{\partial \underline{f}(\underline{x},\underline{u},t)}{\partial \underline{u}}\,\delta\underline{u}.$$

Consequently, when the state variables decrease in magnitude, the multiplier functions, $y_i(t)$, increase in magnitude. That is the two systems are naturally stable in opposite directions. This introduces an extreme sensitivity to the unspecified boundary conditions and effectively rules out methods which iterate on the unknown boundary conditions at one end [69,84].

The solution is obtained through gradient methods in function space which completely decouple the state and adjoint systems and remove the boundary value problem. The basic idea behind these methods lies in that changing a non-optimal control by a small amount given by

$$\delta\underline{u}(t) = \varepsilon \left(\frac{\partial H}{\partial \underline{u}}\right)^T,$$

always improves the objective function by virtue of Eq.(3.11). Several variations of these methods are available [69,70,85]. The method used here is a simple first order one described below:

Step 1. Guess a nominal control profile, $\underline{u}(t)$.

Step 2. Integrate the state system

$$\dot{\underline{x}} = \underline{f}(\underline{x},\underline{u},t), \quad \underline{x}(t_o) \text{ given,}$$

forward from t_o to t_f. Record the resulting $\underline{x}(t)$.

Step 3. Integrate the adjoint system

$$\dot{\underline{y}}^T = \frac{\partial f_o(\underline{x},\underline{u},t)}{\partial \underline{x}} - \underline{y}^T \frac{\partial \underline{f}(\underline{x},\underline{u},t)}{\partial \underline{x}}, \quad \underline{y}^T(t_f)=o$$

backwards from time t_f to t_o.

Record the vector

$$\frac{\partial H}{\partial \underline{u}} = - \frac{\partial f_o(\underline{x},\underline{u},t)}{\partial \underline{u}} + \underline{y}^T \frac{\partial \underline{f}(\underline{x},\underline{u},t)}{\partial \underline{u}}$$

Step 4. Estimate an improved control profile from

$$\underline{u}(t) = \underline{u}(t) + \varepsilon\left(\frac{\partial H}{\partial \underline{u}}\right)^T,$$

where ε is a small positive constant when the resulting $\underline{u}(t)$ does not violate a constraint, and is set to zero otherwise. Repeat steps 2 to 4 until no further improvement in the objective function is obtained.

The positive constant ε represents the step length or gain of the procedure. If it is chosen too small very slow convergence is obtained. If on the other hand it is too large

instability may occur. This can be seen from Figure 3.1, where the successive control profiles obtained with an extremely large ε are shown. In this case, irrespective of the starting profile, the algorithm ends up oscillating between the profiles shown for iterations 19 and 20. To overcome this problem, the gain of the algorithm is made variable by performing a linear search for the largest value of ε which gives an improved performance.

For the problem at hand the Hamiltonian (Eq.3.31) is linear in the control variables μ_1 and μ_2 and

$$\frac{\partial H}{\partial u_j} = wy_j \ , \ j=1,2$$

the optimal controls must therefore switch between their extreme values according to the sign of y_1 and y_2 respectively. However, uncertainties can arise if y_1 and/or y_2 settle at a value of zero,

$$\text{i.e.} \quad \frac{\partial H}{\partial u_j} = o, \text{ over a finite time interval, } j=1,2.$$

The problem is then singular and although the necessary conditions are fulfilled no decision can be made regarding the optimal value of the control variables: the singular controls obtained in this case may or may not be optimal.

In the empirical approaches employed in [46] and [75], the simulated system was forced with a chosen periodic input;

the simulation was continued until the periodicity of the output was established; the performance criteria could then be averaged over one period.

Here we make no assumption of periodicity; our approach is to choose a total operating time that is far in excess of the process settling time, so that contributions to the performance criteria from the start up and shut down transients can be assumed negligible. The only assumption made regarding the form of the input is that it is bounded.

3.2.5. Single control variable

For this case, the state and adjoint systems and the Hamiltonian (Eqs.3.27 to 3.31) can be slightly modified by replacing u_1 and u_2 by the single control variable u: the inlet concentrations are thus switched in phase.
Optimal control policies were obtained for various parameter values and a range of flow rate. Similar behaviour was obtained with any given set of parameters: for flow rates below a critical value $w<w_c$, the optimal dynamic operation corresponded to a fully on-off unimodal periodic input of the type found empirically in chapter 2. However, for $w>w_c$ the method rapidly converged to a steady operation at the maximum allowable concentration level, $u=1$. Figure 3.2 reflects the results obtained for a range of parameters, and a typical iterative progression from an initial steady control policy to a fully developed on-off control is shown

in Figure 3.3. In all cases examined the results obtained were independent of the initial input profile.

3.2.6. Two control variables

Equations (3.27) to (3.31) now apply as they stand: the two reactant concentrations are thus quite independent of each other. In this case, in contrast to the previous one, the results for all flow rates are of the same form: the input concentration of S_1,u_1, takes on its maximum allowable value and that of S_2,u_2, rapidly converges on to an intermediate steady level that depends on the flow rate parameter. Figure 3.4 shows a typical run with the same initial policy as the single variable case above. Identical results were again obtained regardless of the starting control policies.

It is clear that in this case we are faced with a singular problem and we shall now consider it in more detail.

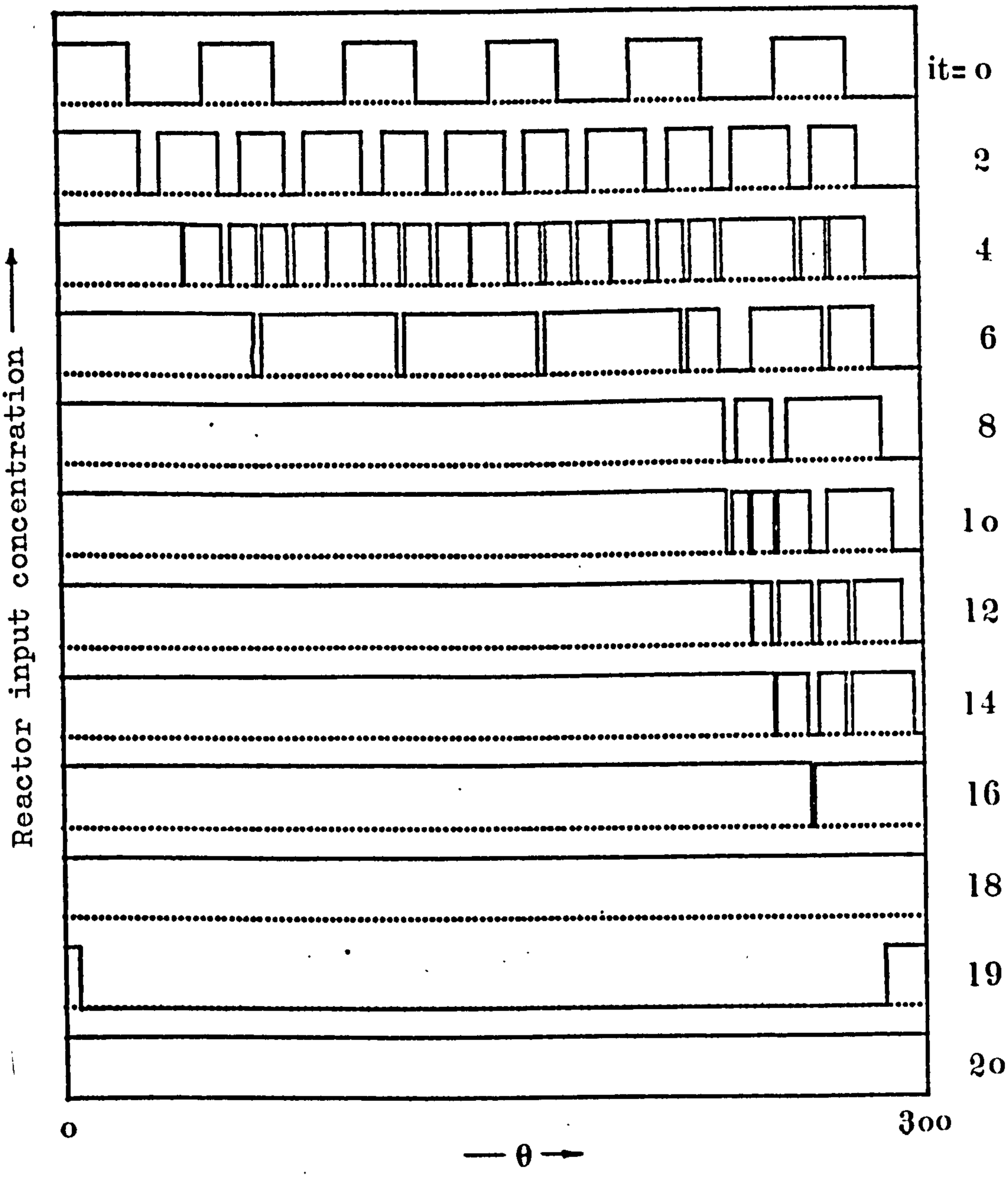


Fig.3.1. Successive input profiles obtained with an infinite gain, $\epsilon = \infty$.
(w=0.25, k_1/k_2=1.0, $k_1\tau_s A_{1fs}$=4.0)

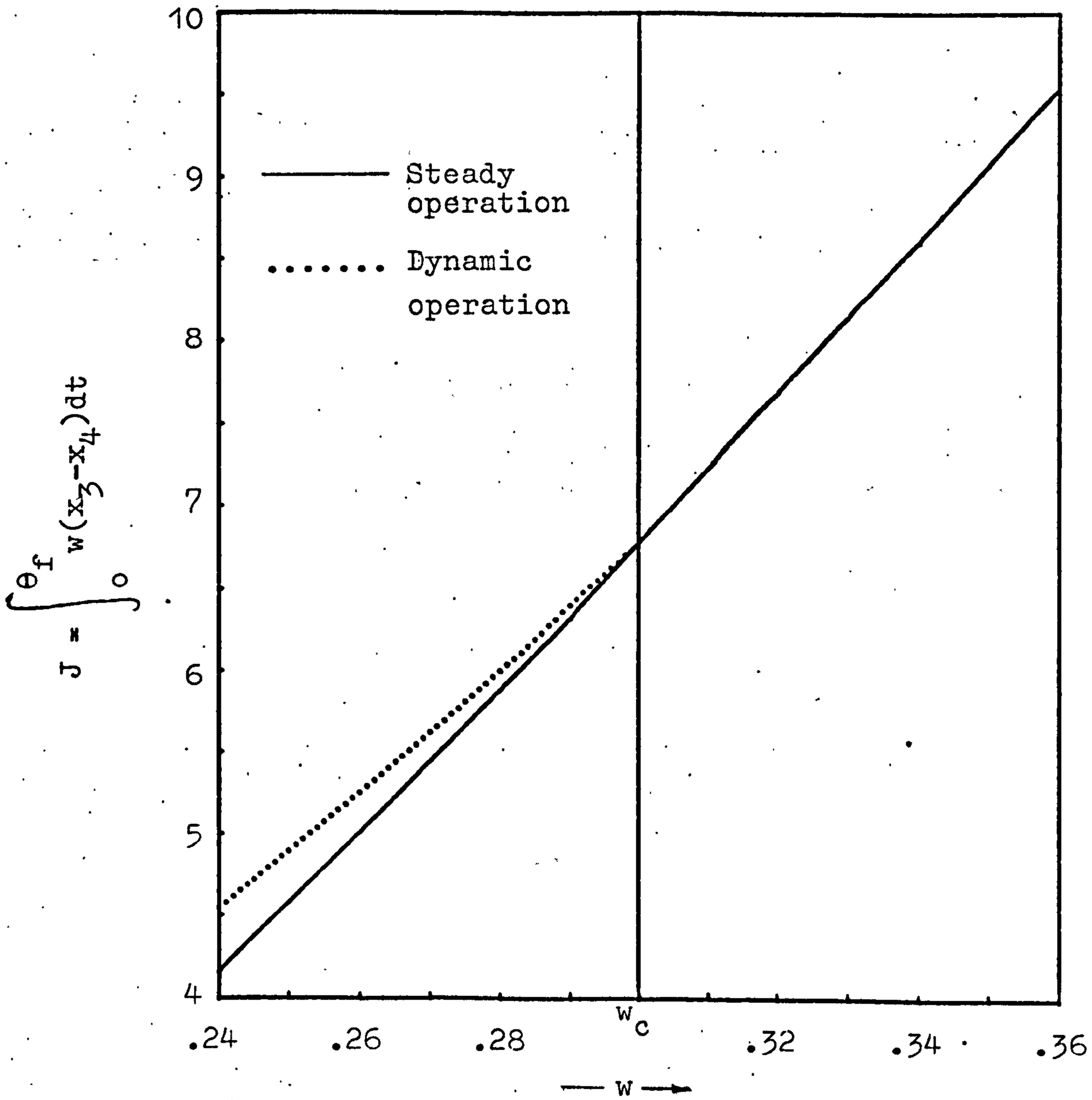


Fig.3.2. Optimal operation of the stirred tank tank reactor for a range of flow rate. For $w < w_c$ dynamic operation is better than steady operation.
($k_1/k_2=1.0$, $k_1\tau_s A_{1fs}=4.0$).

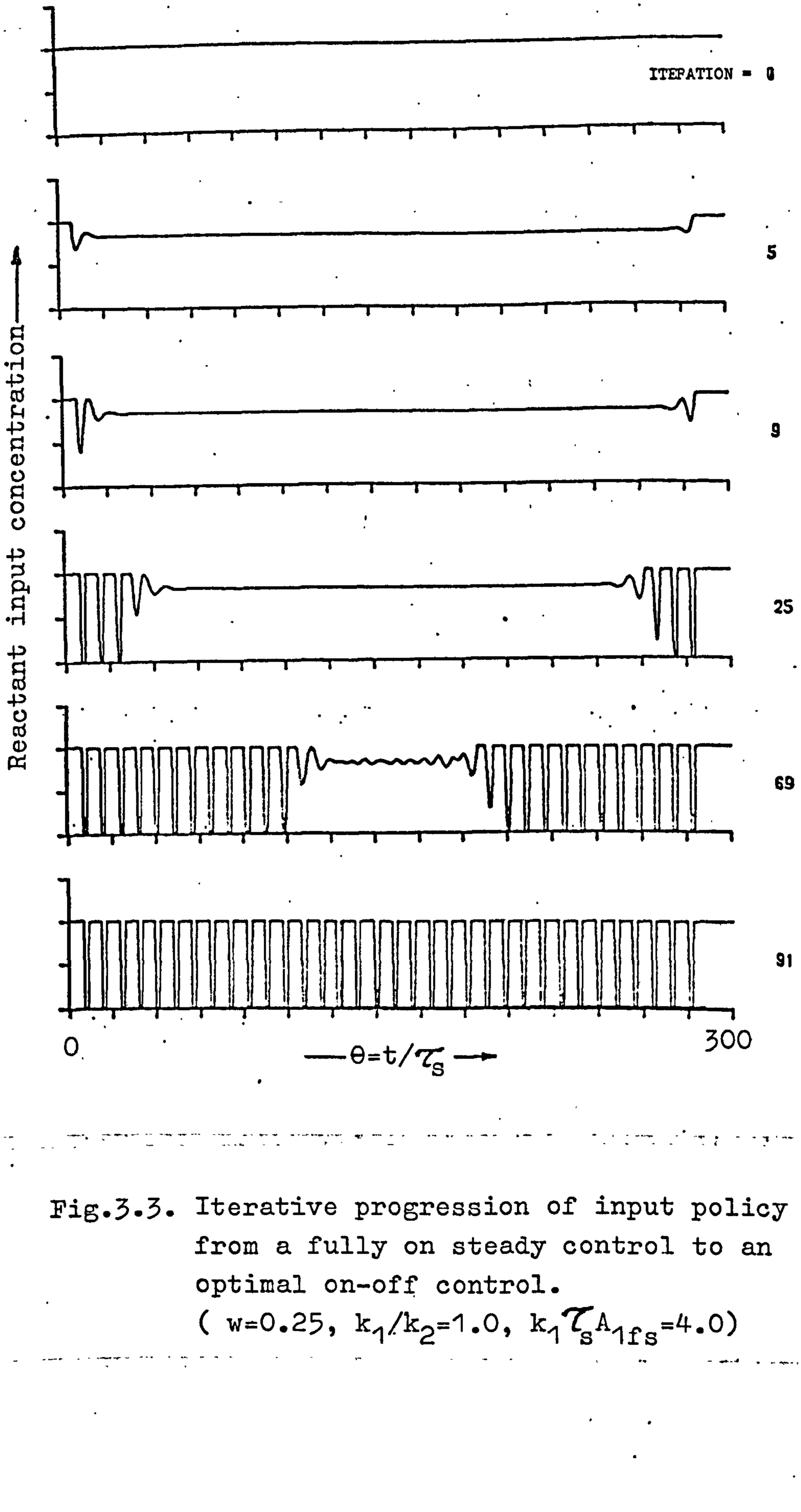


Fig.3.3. Iterative progression of input policy from a fully on steady control to an optimal on-off control.
($w=0.25$, $k_1/k_2=1.0$, $k_1\tau_s A_{1fs}=4.0$)

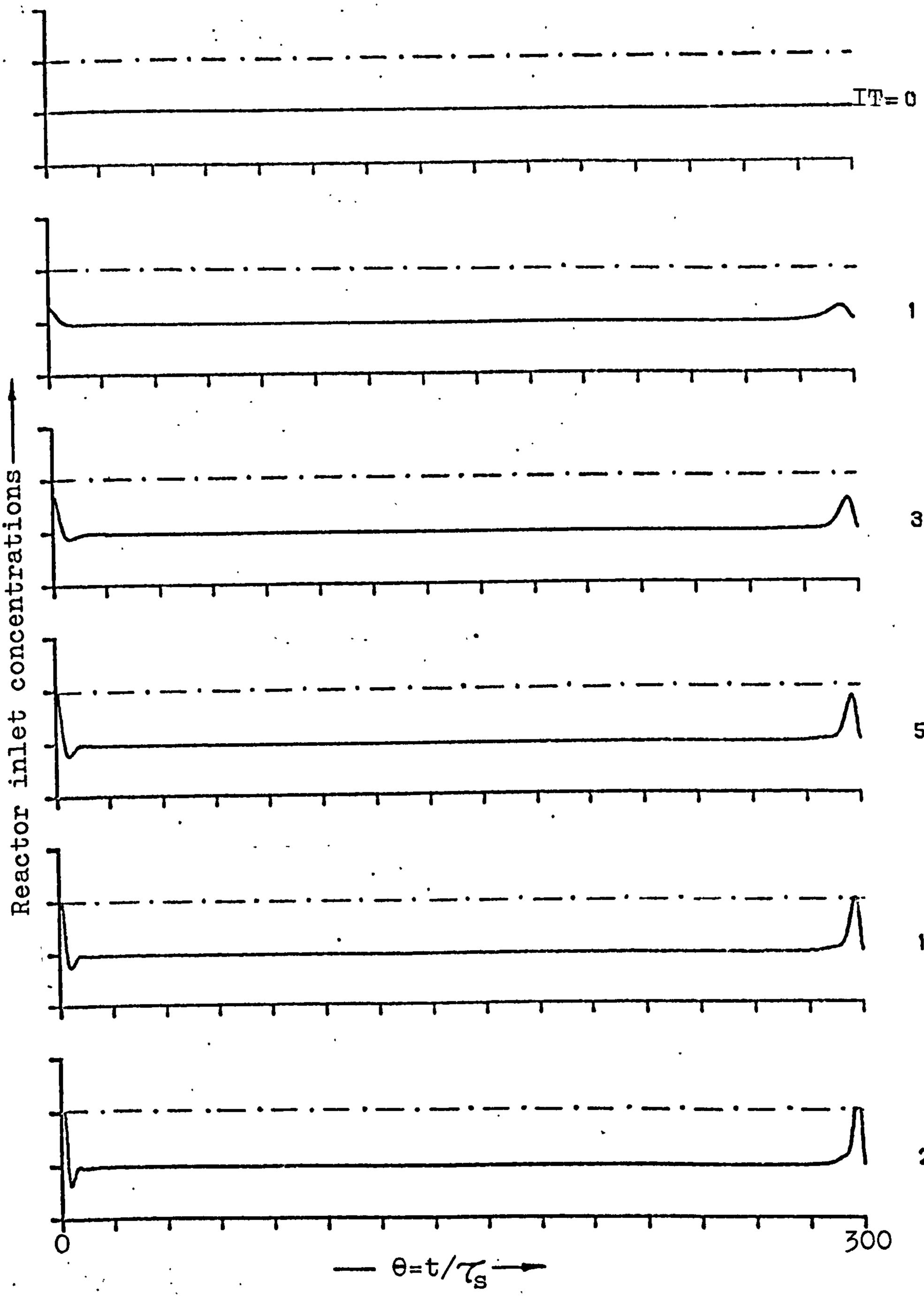


Fig.3.4. Iterative development of a singular control when the inlet concentrations are allowed to vary independently.(-·- u_1, — u_2, w=0.25, k_1/k_2=1.0, $k_1\tau_s A_{1fs}$=4.0)

3.2.7. Singular problems

The necessary condition for optimality of a control vector, $\underline{u}(t)$, is that it maximises the Hamiltonian at almost all points in time. In problems such as ours, the Hamiltonian is composed of linear terms in the control variables and linear and/or nonlinear terms in the state variables. The optimality condition then implies that the control variables assume their extreme admissible levels according to the sign of their respective coefficients, with further improvements not being possible without the violation of the admissible control range.

For the problem in hand inspection of the Hamiltonian (Eq.3.31) reveals that we must have

$$u_j(t) = \begin{cases} u_j^{max} & , \text{ if } y_j(t) > o \\ & , \ j=1,2 \\ u_j^{min} & , \text{ if } y_j(t) < o \end{cases}$$

and

$$y_j(t)\delta u_j(t) \leqslant o \ , \ j=1,2$$

for any admissible variations $\delta u_j(t)$.

However, it may be possible to find a control function, $u_j(t)$, inside the bounded region such that its coefficient in the Hamiltonian is identically zero over a finite time interval. Such controls are called singular and are not determined by the necessary condition that H be at its maximum level.

Instead, a singular control is determined by the fact that its coefficient in the Hamiltonian remains at zero over a finite time interval.

Whenever an extremal control occurs within a bounded region so that

$$\frac{\partial H}{\partial u_j} = o, \tag{3.32}$$

a necessary condition for the optimality of this contol is that

$$\frac{\partial^2 H}{\partial u_j^2} \leqslant o. \tag{3.33}$$

For singular problems Eq. (3.33) is satisfied but yields no information. The question of optimality of a singular control must therefore be answered through additional tests.

A more useful necessary optimality condition for singular controls has been derived [69,85,86] which can be stated as

$$(-1)^m \frac{\partial}{\partial u_j} \left(\frac{d^{2m}}{dt^{2m}} \frac{\partial H}{\partial u_j}\right) \geqslant o, \quad m=o,1,2,\ldots \tag{3.34}$$

For the two control variable case above, over the singular portion of the control u_2

$$\frac{\partial H}{\partial u_2} = wy_2 = o. \qquad 3.35$$

By implicit differentiation of Eq. (3.35) it can be shown that the equations obtained by successive differentiations,

$$\frac{d^k}{dt^k}\frac{\partial H}{\partial u_2} = o, \quad k=1,2,3$$

are independent of u_2. However, on the fourth differentiation we arrive at (see Appendix 3)

$$\frac{1}{w^2}\frac{d^4}{dt^4}\frac{\partial H}{\partial u_2} = S(\underline{x},\underline{y})u_2 + T(\underline{x},\underline{y}) = o, \qquad 3.36$$

where

$$S(\underline{x},\underline{y}) = -w[\alpha_1((\alpha_1 + 2\alpha_2)x_1 - \alpha_1 a) - \alpha_2(2\alpha_2 x_3 - \alpha_1 b)],$$

$$T(\underline{x},\underline{y}) = -\alpha_1\dot{x}_1(w+(\alpha_1+2\alpha_2)x_2)$$

$$+ 2\alpha_2\dot{x}_3(w + \alpha_2 x_2) + \alpha_1\dot{a}(w + \alpha_1 x_2)$$

$$- \alpha_1\alpha_2 x_2\dot{b} - \frac{1}{w}(wx_2 + \alpha_1 x_1 x_2 + \alpha_2 x_2 x_3)S(\underline{x},\underline{y}),$$

Equation (3.36) defines a control law

$$u_2 = -T(\underline{x},\underline{y}) \;/\; S(\underline{x},\underline{y}) \qquad 3.37$$

which a singular control, u_2, must obey. The necessary condition indicated by Eq. (3.34) can now be written as

$$(-1)^2 \frac{\partial}{\partial u_2} \left(\frac{d^4}{dt^4} \frac{\partial H}{\partial u_2}\right) = S(\underline{x},\underline{y}) \leqslant o. \qquad 3.38$$

For all cases where a singular control occured, numerical calculations revealed that Eq. (3.38) is satisfied. However, the singular control predicted by Eq. (3.37) did not correspond to that obtained from the iterative algorithm. This appears to be the result of computational inaccuracy. Equation (3.36) is derived by assuming that $\frac{\partial H}{\partial u_2}$ is exactly zero over the singular control. Whereas, in the iterative algorithm $\frac{\partial H}{\partial u_2}$ oscillates between + 10^{-4} and -10^{-4} and never becomes exactly zero.

Several runs were then made using different storage intervals, initial control policies and operation intervals. In all cases, when the operation interval was sufficiently long the same singular control was obtained. All attempts at forcing the control away from this singularity failed and the algorithm always produced the same result. Consequently, although not proven rigorously, it appears that the singular controls obtained are in fact optimal.

By way of a final example, the results for a short operating interval are given in figure 3.5. In this case, the singular

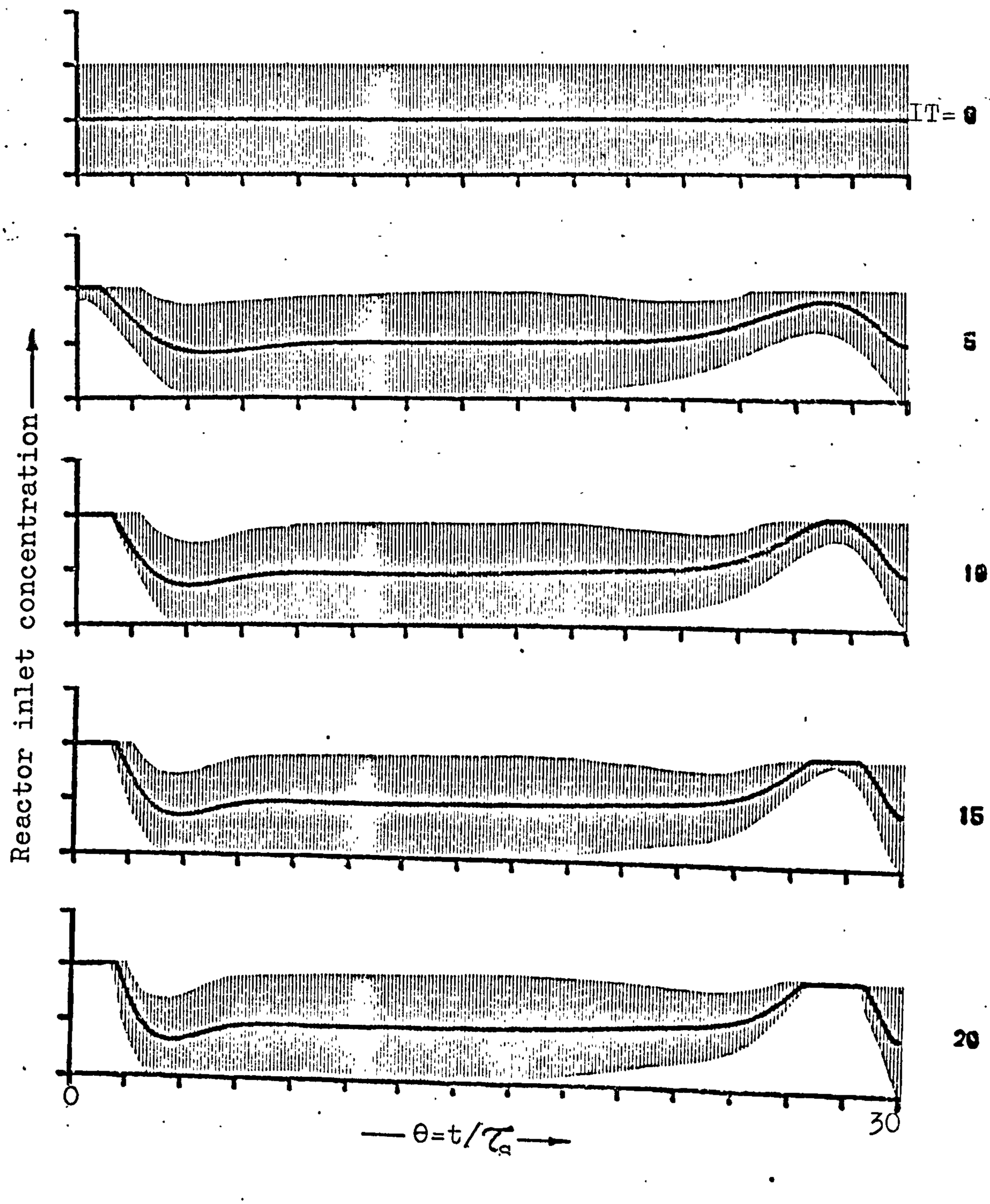


Fig.3.5. Iterative development of a time variable singular inlet concentration for reactant S_2, u_2. The inlet concentration of reactant S_1, u_1, remains at its maximum value, $u(\Theta)=1.0$. (—— steady input, ||| fast switching input, $w=0.25$, $k_1/k_2=1.0$, $k_1\tau_s A_{1fs}=4.0$).

control is not a steady one and varies with time. The same figure shows the results obtained with a rapidly switching initial control policy. It can be seen that the average of the final results obtained are identical to that obtained with a steady starting profile. This goes a long way in confirming the validity of the integration and discretisation procedure used.

3.2.8. Discussion

For all cases tested in which sufficient operating time was allowed for the results to relate to continuous operation, the optimum policies turn out to be rather simple: either steady state or on-off unimodal periodic. The underlying physical reasons for such policies are contained in the consecutive-competing nature of the reactions examined:

$$S_1 + S_2 \longrightarrow S_3,$$
$$S_3 + S_2 \longrightarrow S_4.$$

When the inlet concentration of S_1 and S_2 are constrained to vary together the deciding factor is the mean residence time of the tank. For large volumetric throughputs the concentration of the desired product, S_3, remains sufficiently small. The optimal policy is therefore obtained by keeping the inlet concentrations as high as possible and so promoting the first reaction. However, as the flow rate is reduced the concentration of S_3 is increased to a point where by keeping a high inlet

concentration of S_2 the second reaction is favoured. In such cases it is more profitable to shut off the flow of reactants and flush out the tank until such time that the concentration of S_3 is once again sufficiently small. The optimal policy thus turns out to be on-off periodic. When the inlet concentrations are allowed to vary independently, the first reaction is promoted by a high inlet concentration of S_1. However, the feed concentration of S_2 must be such that the rate of first reaction is at all times larger than that of the second reaction. Thus irrespective of the flow rate used, the optimal policy is a steady operation with the highest possible feed concentration of S_1 and an intermediate steady input concentration of S_2.

The objective function (3.28) would appear to represent the simplest expression of reactor performance that leads to non-trivial solutions when constraints of the type employed in [46] and [75], which stipulate equal average reactant feed rate, are lifted. Production rate is maximised by continuous operation at the maximum permissible levels, but at the expense of selectivity which is favoured by conditions yielding vanishingly small production rates. The objective employed here provides a sensible compromise between these otherwise conflicting performance criteria.

CHAPTER 4

RATIO-INTEGRAL OBJECTIVE FUNCTIONS IN THE OPTIMAL OPERATION OF CHEMICAL REACTORS.

4.1. Introduction

The performance of chemical reactors is generally measured in terms of the overall yield or selectivity of a desired product; these, and indeed the efficiency of most continuous processes must in general be expressed as a ratio of two integral quantities

$$J = \frac{\int_o^{t_f} f_\ell(\underline{x},\underline{u})\,dt.}{\int_o^{t_f} f_m(\underline{x},\underline{u})\,dt} \quad . \qquad 4.1$$

In steady operations the integrands in Eq. (4.1) assume a constant value, so that the objective becomes a simple ratio of two numbers. The steady state overall yield and selectivity are therefore equal to their instantaneous values. In dynamic operations where the form of the inputs are known beforehand, it is often possible to reduce the ratio integral objective to an ordinary integral one. For instance the yield of a desired product is expressed as

$$\eta = \frac{\int_o^{t_f} FA_d\,dt}{\int_o^{t_f} FA_{fk}\,dt}$$

where F is the flow rate, A_d is the exit composition of the desired product and A_{fk} is the inlet concentration of a

key reactant. Then, if the inlet concentration profile of the key reactant is known, the integration in the denominator can be performed beforehand which results in an ordinary integral objective.

In general, however, the dynamic efficiency must be taken as a ratio-integral objective. Furthermore, to satisfy the criteria for strictly comparable conditions for the different modes of operation, it is often necessary to introduce integral side constraints of the type

$$\int_o^{t_f} f_p(\underline{x},\underline{u})dt = M, \text{ a set value} \qquad 4.2$$

In a reactor problem Eq. (4.2) may represent either the total amount of a key component reaching the reactor, or the total production of a certain product.

The approach taken by Bailey and Horn [48,61,62] is to associate an average production rate to each reaction component.

$$\bar{r}_i = \lim_{T\to\infty} \frac{1}{T} \int_o^T r_i(t)dt,$$

where $r_i(t)$ represents the instantaneous production rate of the i^{th} component. The collection of all the points, $\bar{r}_i, i=1,\ldots,n$, which can be achieved with a particular dynamic operation defines a set of points which they term as the attainable set. A dynamic operation is then said to be

improving if this set contains points which cannot be obtained by any steady operation. Our approach differs in as far as, no restrictions about the mode of operation are envisaged and an objective which in effect represents a point efficiency is used.

Our purpose is to demonstrate how objective functions, J, of the above type may be included in the formulation of the problem for solution using the Maximum Principle. The method we propose, as well as being very simple, circumvents the problem of discontinuities often encountered with ratio-type objective functions, and is easily extended to cover additional integral side constraints of the form given by Eq.(4.2). As far as we are aware no previous treatment of this ratio-integral objective function problem has appeared in the literature.

4.2. The basic problem

Briefly we are concerned with the stationary system of state equations

$$\dot{x}_i = f_i(x_1,\ldots,x_n,u_1,\ldots,u_r) = f_i(\underline{x},\underline{u}),$$

$$x_i(o) \text{ given, } i=1,\ldots,n, \qquad 4.3$$

the object being to find an admissible control $\underline{u} = \underline{u}^*(t)$ which, in transferring the phase point from a position $\underline{x}(o)$ to some position $\underline{x}(t_f)$ on a given smooth manifold, Ω, of the state space, minimises the integral functional

$$J = \int_o^{t_f} f_o(x,u)dt. \qquad 4.4$$

The addition of a state variable, x_o, defined through the further state equation

$$\dot{x}_o = f_o(\underline{x},\underline{u}) \ , \ x_o(o) = o \qquad 4.5$$

provides a system of equations which, on integration from time zero to t_f, yields a value of the objective function, J, for any chosen control $\underline{u}(t)$:

$$J = x_o(t_f).$$

The Maximum Principle formulation then involves the definition of adjoint variables, y_i, through the system of equations

$$\dot{y}_o = o,$$

$$\dot{y}_1 = - \frac{\partial f_o(\underline{x},\underline{u})}{\partial x_i} \ y_o - \sum_{j=1}^{n} \frac{\partial f_j(\underline{x},\underline{u})}{\partial x_i} \ y_j \ , \ i=1,\ldots,n, \qquad 4.6$$

and the Hamiltonian function, $H(\underline{x},\underline{y},\underline{u},y_o)$, given by

$$H(\underline{x},\underline{y},\underline{u},y_o) = f_o(\underline{x},\underline{u})\ y_o + \sum_{j=1}^{n} f_j(\underline{x},\underline{u})y_j \qquad 4.7$$

It is then a necessary condition of optimality that y_o be a non-positive constant and the boundary conditions on remaining variables are determined through the transversality conditions [68] which require that the vector $\underline{y}(t_f)=(y_1(t_f),\ldots,y_n(t_f))$ be orthogonal to the tangent plane of manifold Ω passing through the point $\underline{x} = \underline{x}(t_f)$.

The optimal control, $\underline{u}^*(t)$, must then be chosen such that, upon simultaneous solution of the state system (Eqs. 4.3 and 4.5) and the adjoint system (Eqs. 4.6) subject to the boundary conditions dictated by the particular problem at hand, the function $H(\underline{x}(t),\underline{y}(t),\underline{u},y_o)$ viewed as the function of the control, $\underline{u}$, alone, attains its largest possible value for $\underline{u} = \underline{u}^*(t)$ at all times t, $o \leqslant t \leqslant t_f$.

Furthermore, as the adjoint system (Eqs. 4.6) and the Hamiltonian (Eq. 4.7) are linear in the variables y_i, i=1,...,n, the adjoint variables are defined only up to a common multiple. In particular when the manifold Ω coincides with the entire state space, for a non-trivial solution to the adjoint system, it is necessary that

$$y_o < o \tag{4.8}$$

$$y_i(t_f) = o \ , \ i = 1,\ldots,n \tag{4.9}$$

and y_o may be taken as any arbitrary negative constant.

4.3. The ratio-integral objective function

Consider the basic optimal control problem outlined above but with the objective function, J, having the form given by Eq. (4.1) rather than Eq. (4.4). This case can be dealt with by the definition of two further state variables x_ℓ and x_m through:

$$\dot{x}_\ell = f_\ell(\underline{x},\underline{u}), \ x_\ell(o) = o \tag{4.10}$$

$$\dot{x}_m = f_m(\underline{x},\underline{u}), \ x_m(o) = o \tag{4.11}$$

Equation (4.5) now becomes

$$\dot{x}_o = f_o(\underline{x},\underline{u}) = (f_\ell(\underline{x},\underline{u})\ x_m - f_m(\underline{x},\underline{u})\ x_\ell)/x_m^2 \tag{4.12}$$

and the corresponding adjoint variables and their boundary conditions, for this case where the manifold Ω coincides with the entire state space, may be written

$$\dot{y}_\ell = !f_m(\underline{x},\underline{u})y_o/x_m^2 \ , \ y_\ell(t_f) = o \tag{4.13}$$

$$\dot{y}_m = (f_\ell(\underline{x},\underline{u})x_m^2 - 2f_m(\underline{x},\underline{u})\ x_m\ x_\ell)\ y_o/x_m^4, y_m(t_f) = o \qquad 4.14$$

$$\dot{y}_o = o,\ y_o \text{ a negative constant} \qquad 4.15$$

It is noteworthy that Equations (4.13) and (4.14) integrate directly to yield

$$y_\ell = -y_o/x_m + C_\ell \qquad 4.16$$

$$y_m = x_\ell y_o/x_m^2 + C_m \qquad 4.17$$

The constants of integration C_ℓ and C_m are readily obtained from the boundary conditions at time t_f:

$$C_\ell = y_o/x_m(t_f) \qquad 4.18$$

$$C_m = -x_\ell(t_f)y_o/x_m^2(t_f) \qquad 4.19$$

More significantly it is easily demonstrated that the closed form solutions obtained for y_ℓ and y_m (Eqs. (4.16) and (4.17)) lead to the elimination of the discontinuities brought about by the ratio-type nature of the objective function. The adjoint system is now defined through Eqs. (4.15), (4.16) and (4.17) and the system of equations:

$$\dot{y}_i = - \frac{\partial f_o(\underline{x},\underline{u})}{\partial x_i} y_o - \frac{\partial f_\ell(\underline{x},\underline{u})}{\partial x_i} y_\ell - \frac{\partial f_m(\underline{x},\underline{u})}{\partial x_i} y_m$$

$$- \sum_{j=1}^{n} \frac{\partial f_j(\underline{x},\underline{u})}{\partial x_i} y_j \ , \ y_i(t_f) = o \ , \ i=1,\ldots,n \qquad 4.20$$

where $f_o(\underline{x},\underline{u})$ is, by definition, the right hand side of Eq.(4.12). It should be noted that for a physically significant problem it must be assumed that $x_m(t_f) \neq o$. The first three terms in Eqs. (4.20) are discontinuous whenever $x_m(t) = o$, for instance at $t = o$. However these discontinuous terms cancel out with each other, as can be easily verified by substitution for, $f_o(\underline{x},\underline{u}), y_\ell$ and y_m : equations (4.20) then take the form

$$\dot{y}_i = - \frac{\partial f_\ell(\underline{x},\underline{u})}{\partial x_i} C_\ell - \frac{\partial f_m(\underline{x},\underline{u})}{\partial x_i} C_m$$

$$- \sum_{j=1}^{n} \frac{\partial f_j(\underline{x},\underline{u})}{\partial x_i} y_j \ , \ y_i(t_f) = o, \ i=1,\ldots,n, \qquad 4.21$$

It also turns out that the extra terms in the expression for the Hamiltonian (see Eq. 4.7) due to the additional variables y_ℓ and y_m, largely cancel out with the term for y_o to give

$$H(\underline{x},\underline{y},\underline{u},C_\ell,C_m) = f_\ell(\underline{x},\underline{u})C_\ell + f_m(\underline{x},\underline{u})C_m + \sum_{j=1}^{n} f_j(\underline{x},\underline{u})\ y_j \qquad 4.22$$

The Hamiltonian and Eqs. (4.21) are thus independent of y_o, y_ℓ and y_m except in so far as the constants C_ℓ and C_m depend on the boundary values of these variables. Furthermore, as the variables y_i are defined only up to a common multiple and y_o does not directly enter into Eqs. (4.21) and (4.22), C_ℓ may be taken as

$$C_\ell = -1 \qquad 4.23$$

Equations (4.21) and (4.22) can now be written as:

$$\dot{y}_i = \frac{\partial f_\ell(\underline{x},\underline{u})}{\partial x_i} - \frac{\partial f_m(\underline{x},\underline{u})}{\partial x_i} C_m - \sum_{j=1}^{n} \frac{\partial f_j(\underline{x},\underline{u})}{\partial x_i} y_j \ ,\ y_i(t_f) = o,\ i=1,\ldots,n \qquad 4.24$$

$$H(\underline{x},\underline{y},\underline{u},C_m) = - f_\ell(\underline{x},\underline{u}) + f_m(\underline{x},\underline{u}) + \sum_{j=1}^{n} f_j(\underline{x},\underline{u})\ y_j \qquad 4.25$$

and finally Eq. (4.19) reduces to

$$C_m = x_\ell(t_f)/x_m(t_f) = J \qquad 4.26$$

and the necessary optimality conditions for the above problem can now be stated as follows.

For the control, $\underline{u}^*(t)$, and the constant, C_m^*, to be optimal it is necessary that upon simultaneous solution of Eqs. (4.3), (4.10), (4.11) and the system described by Eqs. (4.21), the function $H(\underline{x}(t),\underline{y}(t),\underline{u},\dot{C}_m^*)$, viewed as a function of the control, $\underline{u}$, alone, attains its largest possible value for $\underline{u} = \underline{u}^*(t)$ at all times $t, o \leqslant t \leqslant t_f$; furthermore at the final time, t_f, the following constraint must also be satisfied:

$$C_m^* = x_\ell^*(t_f) \;/\; x_m^*(t_f) = J^*. \qquad 4.27$$

This last condition is somewhat unexpected and worthy of comment: the unknown constant, C_m, introduced by the ratio-integral nature of the objective function must, under optimal conditions, take on the value of the objective function J. This relationship is quite fortunate in the sense that for most physical processes some idea of the possible values of J will be available.

4.4. The integral side constraint

Normally, for the physically more significant problems, in addition to a ratio-integral objective function, an integral side constraint, of the form given by Eq.(4.2), must also be considered. This case can be dealt with by the introduction of a further state variable, x_p, defined through

$$\dot{x}_p = f_p(\underline{x},\underline{u}) \ , \ x_p(o) = o \ , \ x_p(t_f) = M \qquad 4.28$$

then, as the right hand sides of the state equations are independent of x_p, the corresponding adjoint variable, y_p, is given by

$$\dot{y}_p = o, \text{ i.e. } y_p = \text{a constant } C_p \qquad 4.29$$

The major difference with the previous section lies in the nature of the boundary conditions. The manifold Ω no longer coincides with the entire state space, and the boundary conditions of the adjoint system are now given by:

$$y_o \leqslant o,$$

$$y_i(t_f) = o, \ i=1,\ldots,n, \qquad 4.30$$

$$y_\ell(t_f) = y_m(t_f) = o;$$

the boundary condition $y_p(t_f)$, and hence C_p, is however undetermined and in its place the boundary condition $x_p(t_f) = M$ is specified.

In much the same way as before the counterparts of Eqs. (4.21) and (4.22) for this case are developed and may be written as:

$$\dot{y}_i = -\frac{\partial f_\ell(\underline{x},\underline{u})}{\partial x_i} C_\ell - \frac{\partial f_m(\underline{x},\underline{u})}{\partial x_i} C_m - \frac{\partial f_p(\underline{x},\underline{u})}{\partial x_i} C_p$$

$$- \sum_{j=1}^{n} \frac{\partial f_j(\underline{x},\underline{u})}{\partial x_i} y_j \ , \ y_i(t_f) = o, \ i=1,\ldots,n \qquad 4.31$$

$$H(\underline{x},\underline{y},\underline{u},C_\ell,C_m,C_p) = f_\ell(\underline{x},\underline{u})C_\ell + f_m(\underline{x},\underline{u})C_p$$

$$+ \sum_{j=1}^{n} f_j(\underline{x},\underline{u})y_j \qquad 4.32$$

Unlike the previous section the possibility of

$$y_o = o \qquad 4.33$$

can no longer be omitted, and is handled in the following manner. In this instance Eqs. (4.18) and (4.19) imply:

$$C_\ell = C_m = o \ ; \qquad 4.34$$

then, as the adjoint variables are defined only up to a common multiple, for a non-trivial solution of the adjoint system (Eqs. 4.31) to exist, C_p must be a non-zero constant and may be taken as

$$C_p = 1$$

One could then proceed to determine whether a solution of the resulting two point boundary value problem, satisfying the boundary condition $x_p(t_f) = M$, exists.

In the normal case, where $y_o < o$, C_ℓ may be taken as

$$C_\ell = -1 \tag{4.36}$$

and Eqs. (4.31) and (4.32) become

$$\dot{y}_i = \frac{\partial f_\ell(\underline{x},\underline{u})}{\partial x_i} - \frac{\partial f_m(\underline{x},\underline{u})}{\partial x_i} C_m - \frac{\partial f_p(\underline{x},\underline{u})}{\partial x_i} C_p$$

$$- \sum_{j=1}^{n} \frac{\partial f_j(\underline{x},\underline{u})}{\partial x_i} y_j \ , \ y_i(t_f) = o \ , \ i=1,\ldots,n \tag{4.37}$$

$$H(\underline{x},\underline{y},\underline{u},C_m,C_p) = - f_\ell(\underline{x},\underline{u}) + f_m(\underline{x},\underline{u}) C_m + f_p(\underline{x},\underline{u})C_p$$

$$+ \sum_{j=1}^{n} f_j(\underline{x},\underline{u}) y_j \tag{4.38}$$

and finally Equation (4.26) remains unchanged.

The addition of an integral side constraint therefore results in the introduction of an unknown constant, C_p, and an associated necessary optimality condition

$$x_p^*(t_f) = M \ ; \tag{4.39}$$

as a result the optimal control, $\underline{u}^*(t)$, and the constants C_m^* and C_p^* must now be chosen such that, in addition to the usual optimality conditions, at the final time, t_f, relations (4.27) and (4.39) are also satisfied.

Extension to problems in which more than one integral side constraint has to be considered is straight forward and for each of the constraints an additional constant with an associated necessary condition, of the type given by Eq. (4.39) is introduced.

4.5. Integral objective function with an integral side constraint

In this section we aim to point out that the ratio-integral objective function problems can be thought of as a generalisation of cases where, in addition to an objective function

$$J = \int_o^{t_f} f_\ell(\underline{x},\underline{u})\ dt, \qquad 4.40$$

an integral side constraint of the form

$$\int_o^{t_f} f_m(\underline{x},\underline{u})dt = M, \text{ a set value} \qquad 4.41$$

has to be considered. The procedure for these problems is well known and involves the addition of a new state variable, x_m, defined through

$$\dot{x}_m = f_m(x,u) \ , \ x_m(o) = o \ , \ x_m(t_f) = M, \qquad 4.42$$

to the basic problem.

Although for problems of this type we are no longer concerned with a ratio-integral objective function, let us combine Eqs. (4.40) and (4.41) into one entry and consider the objective function, J, defined by

$$J = \frac{\int_o^{t_f} f_\ell(\underline{x},\underline{u}) \ dt}{\int_o^{t_f} f_m(\underline{x},\underline{u}) \ dt} = \frac{1}{M} \int_o^{t_f} f_\ell(\underline{x},\underline{u}) \ dt. \qquad 4.43$$

We are therefore considering the case of a ratio-integral objective function in which the denominator is constrained.

This case differs from the truly ratio-integral one, in as much as the boundary conditions are not the same: they are now given by

$$y_o \leqslant o$$

$$y_i(t_f) = o \ , \ i=1,\ldots,n \qquad 4.44$$

$$y_\ell(t_f) = o$$

Note that, whereas for the truly ratio-integral case the final value of y_m was specified, in this instance $y_m(t_f)$ is

free and in its place it is required that $x_m(t_f) = M$.

The development of the counterparts of Eqs. (4.21) and (4.22) remain as before, but Eqs. (4.18) and (4.19) must now be written as

$$C_\ell = y_o / M \qquad 4.45$$

$$C_m = y_m(t_f) - C_\ell x_\ell(t_f) / M \qquad 4.46$$

The special case $y_o = o$ can be dealt with in the same way as before, and for the case $y_o < o$, C_ℓ may be taken as

$$C_\ell = -1;$$

the Hamiltonian and the adjoint system are again given by Eqs. (4.24) and (4.25). Equation (4.27) must however be replaced by

$$C_m^* = y_m^*(t_f) + x_\ell^*(t_f) / M \qquad 4.47$$

and upon substitution for $y_m^*(t_f)$ from Eq. (4.17) relation (4.47) degenerates into

$$x_m^*(t_f) = M \qquad 4.48$$

Thus the condition $C_m^* = J^*$ for the general ratio-integral objective function, degenerates for the case where the denominator is constrained, to the self-evident form of Eq.(4.48)

4.6. A simple illustrative example

For the purpose of illustration consider the isothermal reaction scheme

$$S_1 \xrightarrow{k_1} S_2 \xrightarrow{k_2} S_3 \qquad 4.49$$

taking place in a constant volume stirred tank reactor and subject to the following conditions:

I) The operation is to take no longer than an interval of time, t_f, and is to be carried out under continuous flow conditions with a fixed flow rate, F.

II) The input concentration of reactant S_1, A_{1f}, cannot exceed a certain maximum allowable level, A_{1fs}.

These conditions effectively imply that an adequate supply of the solvent is available for dilution, so that the input concentration of reactant S_1, A_{1f}, can be made to vary between zero and the maximum allowable level, A_{1fs}.

The process may be represented in the following dimensionless form by reference to a steady-state operation with flow rate, F_s and input concentration A_{1fs}:

$$\frac{dx_1}{d\theta} = Wu - Wx_1 - \alpha_1 x_1, \; x_1(o) = o \qquad 4.50$$

$$\frac{dx_2}{d\theta} = - Wx_2 + \alpha_1 x_1 - \alpha_2 x_2, \; x_2(o) = o$$

where

$$x_i = A_i/A_{1fs}, \; \alpha_i = k_i V/F_s \; , \; i = 1,2 \qquad 4.51$$

$$W = F/F_s, \; u = A_{1f}/A_{1fs}, \; \theta = t \; F_s/V$$

Condition II then implies

$$o \leqslant u(\theta) \leqslant 1 \text{ for all } \theta, \; o \leqslant \theta \leqslant \theta_f.$$

4.6.1. Case (a) the ratio-integral objective function

Consider the case where the objective is to maximise the average yield of the intermediate product, S_2, defined by:

$$\bar{n}_2 = \frac{\int_o^{t_f} FA_2 \, dt}{\int_o^{t_f} FA_{1f} \, dt} \qquad 4.52$$

Introducing a negative sign, the objective function which is now to be minimised, becomes in terms of the dimensionless variables:

$$J = \frac{\int_o^{\theta_f} - Wx_2 \, d\theta}{\int_o^{\theta_f} Wu \, d\theta} \qquad 4.53$$

Following the procedure outlined above for the ratio-integral objective function, the state system is given by Eqs. (4.50) augmented by

$$\dot{x}_\ell = -Wx_2, \; x_\ell(o) = o$$

$$x_m = Wu, \; x_m(o) = o \qquad 4.54$$

The basic adjoint equations become:

$$\dot{y}_1 = (W + \alpha_1)y_1 - \alpha_1 y_2, \; y_1(\theta_f) = o$$

$$\dot{y}_2 = (W + \alpha_2)y_2 - W, \alpha y_2(\theta_f) = o \qquad 4.55$$

with the additional adjoint variables, y_ℓ and y_m, given explicitly by Equations (4.16) and (4.17).

The Hamiltonian for this case is then

$$H = W(C_m + y_1) - ((W + \alpha_1)y_1 - \alpha_1 y_2)x_1 - ((W + \alpha_2)y_2 - W)x_2; \qquad 4.56$$

so that the control, $u(\theta)$ that maximises the Hamiltonian, is on-off switching between zero and unity according to the sign of the switching function $W(y_1(\theta)+C_m)$:

$$u(\theta) = \frac{1}{2}\,(1 + \text{sign}(W(y_1(\theta)+C_m))) \qquad 4.57$$

In order that $u(\theta)$ be optimal ($u^*(\theta)$) it is further required that there exists a value of C_m, C_m^*, which satisfies the necessary optimality condition derived above; namely

$$C_m^* = x_\ell^*(\theta_f) \;/\; x_m^*(\theta_f) = J^* \qquad 4.58$$

For linear processes, such as the one we are considering, the adjoint equations are independent of the state variables, so that Eqs. (4.55) may be integrated backwards from the final time θ_f to time zero. The control, $u(\theta)$ (see Eq. 4.57) is now defined for any given value of C_m; a search procedure then furnishes the optimal value, C_m^*, and hence the optimal control, $u^*(\theta)$, for which, on forward integration of the state system (Eqs. 4.50 and 4.54), relation (4.58) is satisfied.

4.6.2. Case (b) the integral side constraint

This time we consider the problem of maximising the selectivity of the intermediate product, S_2, whilst a fixed amount M of reactant, S_1, is to be used.

Thus the objective function may be written

$$J = \frac{\int_o^{\theta_f} - Wx_2 \, d\theta}{\int_o^{\theta_f} W\,(u-x_1)\, d\theta} \qquad 4.59$$

with the side constraint

$$\int_o^{\theta_f} Wu \, d\theta = M, \text{ a set value} \qquad 4.60$$

The state system is now given by Eqs. (4.50) and

$$\dot{x}_\ell = - Wx_2, \; x_\ell(o) = o$$

$$\dot{x}_m = W(u-x_1), \; x_m(o) = o \qquad 4.61$$

$$\dot{x}_p = Wu, \; x_p(o) = o, \; x_p(\theta_f) = M;$$

and the corresponding adjoint equations become

$$\dot{y}_1 = (W + \alpha_1)y_1 - \alpha_1 y_2 + WC_m,\ y_1(\theta_f) = o$$

$$\dot{y}_2 = (W + \alpha_2)y_2 - W,\ y_2(\theta_f) = o \qquad 4.62$$

$$\dot{y}_p = o,\ \text{i.e. } y_p = \text{a constant, } C_p$$

with y_ℓ and y_m given by Eqs. (4.16) and (4.17).

The Hamiltonian, H, and the control, $u(\theta)$, that maximises it, are now:

$$H = W(y_1+C_m+C_p)u - ((W+\alpha_1)y_1 - \alpha_1 y_2 + WC_m)x_1 - ((W+\alpha_2)y_2 - W)x_2 \qquad 4.63$$

$$u(\theta) = \frac{1}{2}\,(1 + \text{sign}(W(y_1(\theta)+C_m+C_p))) \qquad 4.64$$

The solution is now similar to case (a) except that a two dimensional search on C_m and C_p is now required, to find C_m^* and C_p^* such that, in addition to the optimality condition of Eq. (4.64), the additional necessary conditions, Eqs. (4.27) and (4.39) are satisfied.

4.6.3. Case (c) integral objective function with integral side constraint

In this section we consider the problem of maximising the total output of the intermediate reactant, S_2, for a fixed amount, M, of reactant, S_1, reaching the reactor.

As has been demonstrated above, this problem can be formulated in terms of the ratio-integral objective function of case (a) with the condition

$$C_m^* = J^* \qquad 4.65$$

replaced by

$$x_m^*(\theta_f) = M \qquad 4.66$$

We can therefore use the same search procedure on C_m as in case (a) to find the optimal value, C_m^*, such that relation (4.66) is satisfied.

4.6.4. Results

A simple one dimensional search procedure was used for cases (a) and (c) to find values of C_m that satisfied the relevant constraints (Eqs. 4.27) and (4.48) respectively). For case (b) the method of Rosenbrock [84] was used for the two dimensional search on the values of C_m and C_p that satisfied Eqs. (4.27) and (4.39) simultaneously.

The optimal controls and their corresponding switching functions are shown in Fig.4.1 for the three cases: in each case the control switches once, from maximum to zero, in the operating period. Values of the objective function for these optimal on-off modes of operation are given in Table 4.1 where they are compared with the results of the corresponding (i.e. satisfying the same constraints) constant control operations. In all the above cases the reactor is assumed to be initially free of reaction components.

4.7. Discussion

The above examples were chosen purely to illustrate, as painlessly as possible, applications of the above described method to posed physical problems; they are somewhat contrived in that, the process being linear, no improvement over steady-operation is possible and the only reason that different modes of operation produced different results, and an optimal solution existed at all, was that the objective functions discounted the material left in the reactor at the end of the operating period. Thus for cases (b) and (c), where the switch off occured sufficiently early for the contents of the tank to be, (for all practical purposes) flushed out, the 'optimal' results of Table 4.1 correspond to the true steady-state operation, with the same amount of the reactant reaching the reactor, as can be easily verified.

	Value of objective function J		
	Case (a)	Case (b)	Case (c)
Optimal (on-off)	-o.3632	-o.8ooo	-o.4ooo
Corresponding constant control	-o.344o,(u=1)	-o.7298,(u=.5)	-o.3926,(u=.6)

Table 4.1. Comparison of optimal numerical values of objective functions with corresponding constant control conditions: parameter values as given under Fig. (4.1) and reactor assumed to be initially free of reaction components.

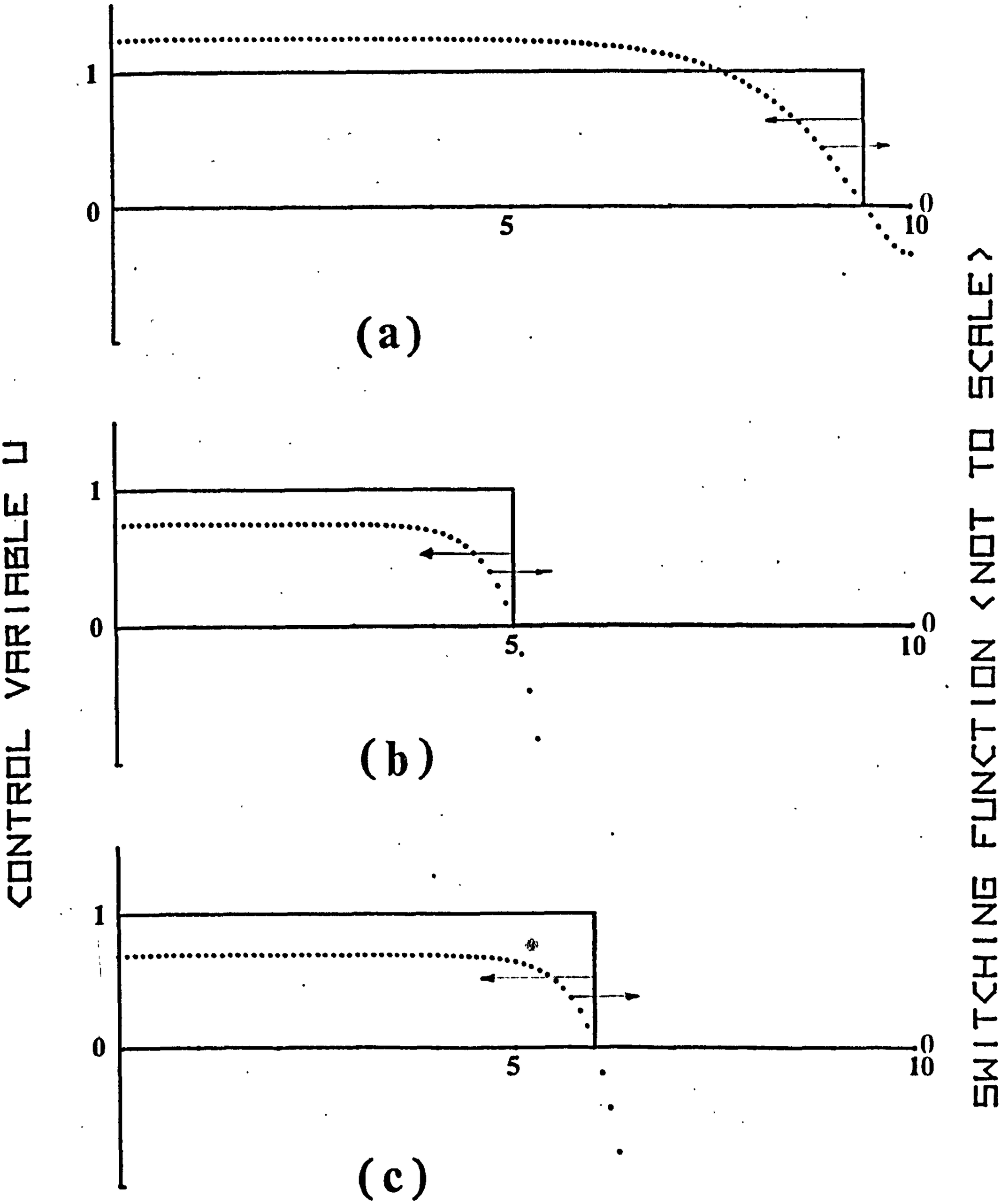


Fig.4.1. Optimal control and switching functions for the illustrative example using the following parameter values: α_1=2.0, α_2=0.5, θ_f=10.0. Case (a): w=0.5; Case (b): w=2.0, M=5.0; Case (c): w=2.0, M=6.0.

However, unsteady-state operation of non-linear processes can lead to significant improvements, in the yield and selectivity of a desired product, over the best possible results under steady-state conditions. An example of this is given by Renken [46] in an empirical study of a consecutive-competing reaction scheme subjected to periodic variations in the reactant feed concentration. It has been further demonstrated [75] that Renken's results are by no means truly optimal and that a rigorous approach of the type we describe above is required.

The order of difficulty of the problem is, however, increased when non-linear processes are considered: as the state and the adjoint systems are naturally stable in opposite directions [69], it may not always be possible to integrate the two systems in the same direction. For linear processes the state and the adjoint systems are independent of each other; it is therefore possible to integrate each one in its naturally stable direction; this is not true of non-linear processes for which the two systems are coupled. Under such circumstances the alternatives open, for the solution of the two point boundary value problems, are either the methods which iterate on the control policy or the state trajectory and which come under the general heading of gradient methods in function space, or the methods which make use of successive linearisation of the non-linear state equations. The development and application of an

efficient algorithm for the solution of problems of this type, arising from realistic chemical processes, will be the subject of the next chapter.

4.8. Conclusion

It has been shown above how objective functions taking the form of the ratio of two integrals may be incorporated into the optimal control problem formulation without giving rise to discontinuities in the system equations.

The formulation resulted in an additional necessary condition of optimality which took the fortunate form

$$C_m = J^* \quad . \qquad 4.67$$

For most problems the possible range of J (and hence the unknown constant C_m) can be narrowed down significantly from physical considerations: for instance, for the reactor problems with which we are primarily concerned, J is bounded by zero and unity and an appropriate first estimate for C_m may be easily arrived at.

Usually comparison between the different modes of operation is only justified under certain conditions: for example in reactor problems a reasonable basis for comparison could be the same amount of key reactants reaching the reactor in all cases; thus it is often necessary to consider integral side constraints; these can be incorporated into the formulation

in the same way as in the usual integral objective function problem; the necessary ratio-integral optimality condition, Eq. (4.67) remains unaffected.

Although we have focused our attention on reactor problems, and in particular on the well mixed continuous flow reactor, the above procedure may equally well be applied to any process (batch or continuous) describable by a system of ordinary differential equations.

CHAPTER 5

DEVELOPMENT OF A GENERAL ALGORITHM FOR DETERMINATION OF OPTIMAL PERIODIC OPERATIONS

The application of variational methods to the determination of optimal unsteady modes of operation often yields valuable qualitative information about the best control strategies. However for reasons already discussed the quantitative determination of these best strategies is not easily accomplished. The aim of this chapter is to lay down the mathematical foundations of a computationally efficient algorithm for the solution of such problems.

While seeking a dynamic operation to improve the efficiency of a continuous process we are faced with two problems. The first difficulty arises from the need to consider ratio-integral objective functions and was dealt with in chapter 4. The second problem is one of continuity: that is to ensure that a dynamic operation obtained over a finite operating interval is in fact comparable with a continuous operation over an infinite length of time. In chapter 3 this problem was overcome by using a very long operating interval so that the start-up and shut-down transients could be neglected. Consequently, the storage area and computation times required for the solution of the optimal control problem were quite large.

An alternative is to look for an optimal <u>periodic</u> control strategy whose repeated application yields an optimal periodic process. In this case one could consider a much shorter interval of a single period with subsequent savings in computational time and storage. This is the approach employed here: the algorithm developed below is a computationally efficient procedure for determination of optimal periodic modes of unsteady operation.

5.1. Numerical solution of optimal control problems

In the majority of applications the solution of an optimal control problem involves finding, in a given time interval, $t_o \leqslant t \leqslant t_f$, the following:

a) the control vector $\underline{u}(t)$, an r-vector function,

b) the state vector $\underline{x}(t)$, an n-vector function,

c) the adjoint vector $\underline{y}(t)$, an n-vector function,

such that

i) the n system differential equations (involving $\underline{x},\underline{u}$) are satisfied,

ii) the n adjoint differential equations (involving $\underline{x},\underline{u},\underline{y}$) are satisfied,

iii) the 2n boundary conditions on $\underline{x}(t)$ and $\underline{y}(t)$ some of which may be given at the initial time, t_o, and others at the terminal time, t_f, are satisfied,

iv) the control constraints, if any, are not violated,

v) the integral side constraints (involving $\underline{x},\underline{u}$), if any, are satisfied,

vi) the r necessary conditions of optimality (involving $\underline{x},\underline{u},\underline{y}$) are fulfilled

This constitutes a complicated two-point boundary-value problem which in all but the trivial cases must be solved numerically. This involves either flooding or iterative procedures. In flooding (or dynamic programming) the unspecified boundary conditions at one end are guessed and the resulting initial value problem is solved. This procedure is repeated until a range of unspecified boundary conditions at this end has been covered. Then provided the correct range is chosen,

some of the trajectories will end near the specified boundary conditions at the other end; thus, providing an approximate solution of the two-point boundary value problem.

In iterative procedures a nominal solution is chosen which satisfies some of conditions (i) through (vi) above. The nominal solution is then gradually modified until all the conditions are fulfilled to the desired accuracy. Variations of three methods have been used, particularly in application to unconstrained control problems. These are the neighbouring extremal, gradient, and quasilinearisation methods which are reviewed in chapter 7 of [69]. There are several algorithms for linear constrained control problems and these are reviewed by Plant [87] who also gives several computer programs.

In the case of unconstrained or linear constrained control problems, it is usually possible to produce criteria for the existence of optimal controls and their uniqeness. In general, this is not possible for nonlinear constrained control problems; as a result the algorithms for this type of problem tend to be rather specific and problem orientated.

The algorithm developed resembles that given by Horn and Lin [47], in as far as the periodicity condition forms an integral part of it. It differs from their algorithm in the sense that the use of a particular linearisation enables the solution of more complicated problems to be obtained, including those with ratio-integral objectives and integral side constraints.

5.2. The optimal periodic control problem

The problem examined in this chapter is to find a periodic control vector, $\underline{u}(t)$, over a given period t_p,

$$\underline{u}(t+t_p) = \underline{u}(t), \text{ for any } t, \tag{5.1}$$

such that the process of intersest,

$$\dot{\underline{x}} = \underline{f}(\underline{x},\underline{u}), \tag{5.2a}$$

is also periodic,

$$\underline{x}(t+t_p) = \underline{x}(t), \text{ for any } t; \tag{5.2b}$$

a given set of integral constraints,

$$\int_t^{t+t_p} f_i^+(\underline{x},\underline{u})\, dt = M_i, \text{ a set value, } i=1,\ldots,q, \tag{5.3}$$

are satisfied; and a ratio-integral objective function,

$$J = \frac{\int_t^{t+t_p} f_{q+2}^+(\underline{x},\underline{u})\, dt}{\int_t^{t+t_p} f_{q+1}^+(\underline{x},\underline{u})\, dt}, \tag{5.4}$$

is minimised.

In chapter 3 a continuous operation was obtained by choosing a sufficiently lengthy operating interval for the start-up and shut-down effects to be negligible. In the present case a continuous operation is explicitly implied by the periodicity conditions (5.1) and (5.2b). We can therefore focus our attention on a single period and examine equations (5.1-5.4) between t=0 and $t=t_p$. This, as we shall see later, results in enormous savings of computational time and storage.

Introducing a new set of additional state variables defined through

$$\dot{\underline{x}}^+ = \underline{f}^+(\underline{x},\underline{u}),\ \underline{x}^+(0)=0, \qquad 5.5$$

where

$$\underline{x}^+ = (x_1^+, \ \ldots \ , x_{q+2}^+)^T \text{and } \underline{f}^+ = (f_1^+, \ \ldots \ , f_{q+2}^+)^T,$$

the maximum principle formulation of this problem remains as that given in chapter 4 with the exception of the boundary conditions on the adjoint system

$$\dot{\underline{y}} = - \left(\frac{\partial \underline{f}^+}{\partial \underline{x}}\right)^T \underline{c} - \left(\frac{\partial \underline{f}}{\partial \underline{x}}\right)^T \underline{y}, \qquad 5.6a$$

which by virtue of the periodicity condition (5.2b) must now be stated as

$$\underline{y}(0) = \underline{y}(t_p). \qquad 5.6b$$

The optimal control vector, $\underline{u}^*(t)$, and the optimal constant vector $\underline{c}^*=(c_1^*,\ldots,c_{q+2}^*)$ must then be such that the

differential systems (5.2) and (5,6) are satisfied; the Hamiltonian

$$H(\underline{x},\underline{u},\underline{y},\underline{c}) = \underline{c}^T \underline{f}^+(x,\underline{u}) + \underline{y}^T \underline{f}(\underline{x},\underline{u}), \qquad 5.7$$

attains its largest possible value at the point $\underline{u}=\underline{u}^*(t)$; and the integral side constraints are satisfied

$$x_i^{+*}(t_p) = M_i, \text{ a set value, } i=1,\ldots,q. \qquad 5.8a$$

It also turns out that as in chapter 4 we may take $c^*_{q+2}= -1$, and the optimal value of c_{q+1} must then satisfy

$$c^*_{q+1}= J^* = \frac{x^+_{q+2}(t_p)}{x^{+*}_{q+1}(t_p)} . \qquad 5.8b$$

5.3. The linearised system

For reasons discussed in chapter 3, the simultaneous integration of the state (5.2) and the adjoint (5.6) systems is generally not possible. The results obtained by the decoupled integration of these systems were also found to be extremely sensitive to the boundary conditions. The problem here is further complicated by the need to match the periodicity conditions (5.2b) and (5.6b). To overcome these difficulties we shall solve the optimal control problem for a certain linearised version of the original nonlinear system (5,2). We shall then gradually

modify this linearised system until its solution coincides with that for the nonlinear system.

Let $\underline{u}^k(t)$ be a periodic control vector whose repeated application leads to a periodic phase trajectory $\underline{x}^k(t)$. The linearised system is then obtained by the linearisation of systems (5.2) and (5.5) with respect to the trajectory $\underline{x}^k(t)$:

$$\dot{\underline{z}} = \underline{f}(\underline{x}^k(t),\underline{v}) + \left(\frac{\partial \underline{f}}{\partial \underline{x}}\right)^k (\underline{z} - \underline{x}^k(t)),\ \underline{z}(0)$$

$$\dot{\underline{z}}^+ = \underline{f}^+(\underline{x}^k(t),\underline{v}) + \left(\frac{\partial \underline{f}^+}{\partial \underline{x}}\right)^k (\underline{z} - \underline{x}^k(t)),\ \underline{z}^+(0) \qquad 5.9$$

where $\frac{\partial \underline{f}}{\partial \underline{x}}$ is a n x n matrix, $\frac{\partial \underline{f}^+}{\partial \underline{x}}$ is a (q+2) x n matrix, the superscript k denotes evaluation with $\underline{u}^k(t)$ and $\underline{x}^k(t)$, and $\underline{v}=\underline{v}(t)$ is an r-vector of as yet undetermined control variables. The choice of the control $\underline{v}(t)$ and the initial state $\underline{z}(0)$ then determines the state trajectory, $\underline{z}(t)$, for the linearised system. System (5.9) is closely related to the nonlinear systems (5.2) and (5.5). It can be easily shown that the adjoint system for a process described by (5.9) is identical to to that for the original nonlinear system. Another property of the linearised system comes to light if we subtract the original nonlinear system from it. Considering the first equation in (5.9) we have:

$$\frac{d}{dt}(\underline{z}(t)-\underline{x}^k(t)) = \underline{f}(\underline{x}^k(t),\underline{v}(t)) - \underline{f}(\underline{x}^k(t),\underline{u}^k(t)) + \left(\frac{\partial \underline{f}}{\partial \underline{x}}\right)^k (\underline{z}(t)-\underline{x}^k(t)),\ \underline{z}(0)-\underline{x}^k(0)$$

Now, if we choose $\underline{v}(t)=\underline{u}^k(t)$ and $\underline{z}(0)=\underline{x}^k(0)$ the above equation reduces to a homogeneous system of linear differential equations

$$\frac{d}{dt}(\underline{z}(t)-\underline{x}^k(t)) = (\frac{\partial_{\tau}\underline{f}}{\partial \underline{x}})^k \ (\underline{z}(t)-\underline{x}^k(t)), \ (\underline{z}(0)-\underline{x}^k(0))=0,$$

whose general solution is given by

$$\underline{z}(t) - \underline{x}^k(t) = 0.$$

In other words, provided the same control histories, $\underline{v}(t)=\underline{u}^k(t)$, and initial conditions, $\underline{z}(0)=\underline{x}^k(0)$, are used the linearised and nonlinear systems yield identical results.

In the sequel we shall use the above properties such that by successive solution of the optimal control problem for the linearised system, the solution for the nonlinear system is gradually approached. To this end we shall first present a general algorithm for problems linear in the state variables which will form the central core for the solution of nonlinear problems.

5.4. The general solution of system equations for problems linear in the state variables.

Let us assume that the state variables are defined through

$$\dot{\underline{z}} = F(t)\ \underline{z} + \underline{g}(\underline{v},t),\ \underline{z}(0),$$

and 5.10

$$\dot{\underline{z}}^{+} = F^{+}(t)\ \underline{z} + \underline{g}^{+}(\underline{v},t),\ \underline{z}^{+}(0)=0,$$

where $F(t)$ and $F^{+}(t)$ are given n x n and (q+2) x n matrices, and $\underline{g}(\underline{v},t)$ and $\underline{g}^{+}(\underline{v},t)$ are given n-vector and (q+2)-vector functions of the control $\underline{v}(t)$ and time t.

The adjoint system for Eqs.(5.10) is therefore given by

$$\dot{\underline{y}}^{T} = -\underline{c}^{T}\ F^{+}(t) - \underline{y}^{T}\ F(t),\ \underline{y}^{T}(t_p), \qquad 5.11$$

and the Hamiltonian takes the form

$$H(\underline{z},\underline{v},\underline{y},\underline{c}) = \underline{c}^{T}\ [\ F^{+}(t)\ \underline{z} + \underline{g}^{+}(\underline{v},t)\ ] + \underline{y}^{T}\ [\ F(t)\underline{z} + \underline{g}(\underline{v},t)\ ] \qquad 5.12$$

Introducing the partitioned vectors and matrices

$$\underline{Z}^{T}(t) = (\underline{z}^{T}(t),\underline{z}^{+T}(t)),\ \underline{Y}^{T}(t) = (\underline{y}^{T}(t),\underline{c}^{T}),$$

$$\underline{b}^{T}(\underline{v},t) = (\underline{g}^{T}(\underline{v},t),\underline{g}^{+T}(\underline{v},t)),$$

$$A(t) = \begin{bmatrix} F(t) & 0 \\ \hline F^{+}(t) & 0 \end{bmatrix},$$

Eqs.(5.10-5.12) can be written as

$$\frac{d\ \underline{Z}}{d\ t} = A(t)\ \underline{Z} + b(\underline{v},t),\quad \underline{Z}(0) = \begin{bmatrix} \underline{z}(0) \\ \hline 0 \end{bmatrix} \qquad 5.13$$

$$\frac{d\ \underline{Y}}{d\ t} = -\ A^T(t)\ \underline{Y},\quad \underline{Y}(t_p) = \begin{bmatrix} \underline{y}(t_p) \\ \hline \underline{c} \end{bmatrix} \qquad 5.14$$

$$H(\underline{Z},\underline{u},\underline{Y}) = \underline{Y}^T\ A(t)\ \underline{Z} + \underline{Y}^T\ \underline{b}(\underline{v},t) \qquad 5.15$$

Now, the general solution of Eqs.(5.13) and (5.14), which are linear in the state variables, can be written down as a fuction of a single transition matrix $\phi(t)$ (see Appendix 4), in the following form

$$\underline{Z}(t) = \phi^{-1}(t)\ \left[\ \phi(0)\underline{Z}(0) + \int_0^t \phi(t)\underline{b}(\underline{v},t)\ dt\ \right], \qquad 5.16$$

$$\underline{Y}(t) = \phi^T(t)\ \underline{Y}(t_p), \qquad 5.17$$

where $\phi(t)$ is a $(n+q+2)$ x $(n+q+2)$ transition matrix defined through the matrix differential equation:

$$\frac{d}{dt}\ \phi(t) = -\ \phi(t)\ A(t),\ \phi(t_p) = I \qquad 5.18$$

By examining Eq.(5.18) in a partitioned form

$$\frac{d}{dt}\ \left[\begin{array}{c|c} \phi_1 & \phi_3 \\ \hline \phi_2 & \phi_4 \end{array}\right] = -\ \left[\begin{array}{c|c} \phi_1 & \phi_3 \\ \hline \phi_2 & \phi_4 \end{array}\right]\ \left[\begin{array}{c|c} F & 0 \\ \hline F^+ & 0 \end{array}\right],\ \left[\begin{array}{c|c} I_n & 0 \\ \hline 0 & I_{q+2} \end{array}\right],$$

it can be easily shown that

$$\phi(t) = \left[\begin{array}{c|c} \phi_1(t) & 0 \\ \hline \phi_2(t) & I_{q+2} \end{array}\right] , \qquad 5.19$$

and its inverse is given by

$$\phi^{-1}(t) = \left[\begin{array}{c|c} \phi_1^{-1}(t) & 0 \\ \hline -\phi_2(t)\phi_1^{-1}(t) & I_{q+2} \end{array}\right] . \qquad 5.20$$

Substituting the partitioned expressions for $\underline{Z}(t)$, $\underline{Y}(t)$, $A(t)$, $\underline{b}(\underline{v},t)$, $\phi(t)$, and $\phi^{-1}(t)$ into Eqs.(5.13) to (5.18) and simplifying we arrive at:

$$\underline{z}(t) = \phi_1^{-1}(t) \left[\phi_1(0)\underline{z}(0) + \int_0^t \phi_1(t)\underline{g}(\underline{v},t)\, dt \right], \qquad 5.21$$

$$\underline{z}^+(t) = -\phi_2(t)\underline{z}(t) + \phi_2(0)\underline{z}(0) + \int_0^t \left[\phi_2(t)\underline{g}(\underline{v},t) + \underline{g}^+(\underline{v},t) \right] dt, \qquad 5.22$$

$$\underline{y}(t) = \phi_1^T(t)\underline{y}(t_p) + \phi_2^T(t)\underline{c} , \qquad 5.23$$

$$\underline{c}(t) = \text{a constant vector}, \qquad 5.24$$

$$H = \underline{y}^T(t_p)\phi_1(t)[\ F(t)\ \underline{z}(t) + \underline{g}(\underline{v},t)\ ]$$

$$+ \underline{c}^T[\ F^+(t) + \phi_2(t)F(t)\ ]\ \underline{z}(t)$$

$$+ \underline{c}^T[\ \phi_2(t)\ \underline{g}(\underline{v},t) + \underline{g}^+(\underline{v},t)\ ], \qquad 5.25$$

where $\phi_1(t)$ and $\phi_2(t)$ are defined through

$$\frac{d}{dt}\phi_1(t) = -\ \phi_1(t)F(t),\ \phi_1(t_p)=I_n, \qquad 5.26$$

$$\frac{d}{dt}\phi_2(t) = -\ F^+(t) - \phi_2(t)F(t),\ \phi_2(t_p)=0. \qquad 5.27$$

Equations (5.21) to (5.27) give the general solution of the state and adjoint systems for given initial conditions on the state variables, $\underline{z}(0)$, and given final boundary conditions on the adjoint variables, $\underline{y}(t_p)$.

5.4.1. A necessary condition for periodic operation

When a process is subjected to periodic inputs some time must elapse before the outputs assume a periodic behaviour. For the linear system under consideration the state at the begining and the end of each period, for a given control, $\underline{v}(t)$, are related through (see Eqs.5.21 and 5.26):

$$\underline{z}(t_p) = \phi_1(0)\ \underline{z}(0) + \int_0^{t_p} \phi_1(t)\ \underline{g}(\underline{v},t)\ dt. \qquad 5.28$$

Similarly, the adjoint vectors at the begining and end of each period are related through (see Eq.5.23):

$$\underline{y}(0) = \phi_1^T(0)\ \underline{y}(t_p) + \phi_2^T(0)\ \underline{c}\ . \qquad 5.29$$

Thus, the periodicity condition $\underline{z}(0)=\underline{z}(t_p)$ implies that

$$\underline{z}(t_p) = \underline{z}(0) = [\ I_n - \phi_1(0)\ ]^{-1} \int_0^{t_p} \phi_1(t)\underline{g}(\underline{v},t)dt, \qquad 5.30$$

and the periodicity condition $\underline{y}(0)=\underline{y}(t_p)$ implies that

$$\underline{y}^T(t_p) = \underline{y}^T(0) = \underline{c}^T\ \phi_2(0)\ [\ I_n - \phi_1(0)\ ]^{-1}. \qquad 5.31$$

Clearly, the existence of the inverse of $[\ I_n - \phi_1(0)\ ]$ is a necessary condition for the periodic operation of a linear process. Furthermore, as the adjoint systems for the linearised and nonlinear systems are identical, it is also a necessary one for the nonlinear process.

In other words, the application of a periodic control, $\underline{v}(t)$, leads to a periodic phase trajectory only if

$$\det\ [\ I_n - \phi_1(0)\ ] \neq 0. \qquad 5.32$$

It should not escape our attention that if (5.32) is satisfied, the periodic end state for a process linear in the state variables is directly given by Eq.(5.32): the initial transients in this case are therefore effectively cut out.

5.4.2. An algorithm for periodic operation of processes linear in the state variables.

We shall now formulate the algorithm which will form the central core for solution of nonlinear problems. The final value $\underline{z}^{+}(t_p)$ and the Hamiltonian, H, for a periodic operation with the control vector $\underline{v}(t)$ are easily obtained by substitution of Eqs.(5.30) and (5.31) into (5.22) and (5.25). After some algebraic manipulations we arrive at:

$$\underline{z}^{+}(t_p) = \int_0^{t_p} [\ \beta(t)\ \underline{g}(\underline{v},t) + \underline{g}^{+}(\underline{v},t)\ ]\ dt, \qquad 5.33$$

and

$$H = \underline{c}^{T}[\ \beta(t)\ F(t) + F^{+}(t)\ ]\underline{z}(t) + \underline{c}^{T}[\ \beta(t)\ \underline{g}(\underline{v},t) + \underline{g}^{+}(\underline{v},t)\ ], \qquad 5.34$$

where $\beta(t)$ is a $(q+2) \times n$ matrix given by

$$\beta(t) = \phi_2(0)[\ I_n - \phi_1(0)\ ]^{-1}\phi_1(t) + \phi_2(t). \qquad 5.35$$

The problem is thus reduced to that of finding a set of constants $c_1^*, \ldots, c_{q+1}^*$ (with $c_{q+2}^* = -1$) and a control vector $\underline{v}^*(t)$ such that: the Hamiltonian (5.34) viewed as a function of the control $\underline{v}$ alone achieves its largest possible value for $\underline{v}=\underline{v}^*(t)$ for all t, $0 \leqslant t \leqslant t_p$, and the following relations are satisfied

$$z_i^{+*}(t_p) = M_i, \text{ a set value, } i=1,\ldots,q, \qquad 5.36a$$

$$c_{q+1}^* = J^* = \frac{z_{q+2}^*(t_p)}{z_{q+1}^*(t_p)} \qquad 5.36b$$

Let us combine Eqs.(5.36) into a single augmented objective, $\bar{J}$, defined by

$$\bar{J} = \psi(\underline{z}^{+}(t_p)) = \frac{1}{2}\,[c_{q+1} - J]^2 + \frac{\omega}{2}\sum_{i=1}^{q}[z_i^{+}(t_p) - M_i]^2, \qquad 5.37$$

where ω is a positive weighting scalar. Clearly, $\bar{J}$ has its minimum value when Eqs.(5.36) are satisfied. The problem could then be solved through the following procedure:

Step 1. Guess a set of values for $c_1,\ldots,c_{q+1}$ and put $c_{q+2} = -1$.

Step 2. Determine and store $\phi_1(t)$ and $\phi_2(t)$ by the backward integration of

$$\dot{\phi}_1 = -\phi_1(t)F(t),\ \phi_1(t_p) = I_n,$$
$$\dot{\phi}_2 = -F^{+}(t) - \phi_2(t)F(t),\ \phi_2(t_p) = 0.$$

Step 3. Invert $[I_n - \phi_1(0)]$ and store the matrix $\beta(t)$:

$$\beta(t) = \phi_2(0)[I_n - \phi_1(0)]^{-1}\phi_1(t) + \phi_2(t).$$

Step 4. Find the control vector $\underline{v}^{\circ}(t)$ which maximises the function

$$h = \underline{c}^{T}[\beta(t)\underline{g}(\underline{v},t) + \underline{g}^{+}(\underline{v},t)]$$

at each time t, $0 \leqslant t \leqslant t_p$. Determine

$$\underline{z}^{+\circ}(t_p) = \int_0^{t_p}[\beta(t)\underline{g}(\underline{v}^{\circ};t) + \underline{g}^{+}(\underline{v}^{\circ};t)]\,dt,$$

and evaluate $\bar{J} = \psi(\underline{z}^{+\circ}(t_p))$.

Step 5. Adjust the values of c_i, i=1,...,q+1, and repeat Steps 4 and 5 until no further reduction in $\bar{J}$ can be obtained. The optimal control is then given by $\underline{v}^*(t)=\underline{v}^\circ(t)$.

The only numerical integration involved in the above procedure is carried out once in Step 2. The only matrix inversion necessary is that of $[I_n-\phi_1(0)]$ in Step 3 which provides a test for the possibility of periodic operation. The matrix $\beta(t)$ does not require extra storage space and can be stored in the same location as $\phi_2(t)$. The relative accuracy to which conditions (5.36a) and (5.36b) are satisfied can be adjusted by the appropriate choice of the weighting scalar, ω in Eq.(5.37).

As previously experienced the final value of $\underline{z}^+(t_p)$ is extremely sensitive to the value of the constants c_i,i=1,..,q+1. In some cases the direct inspection of

$$h = \underline{c}^T[\ \beta(t)\ g(\underline{v},t) + g^+(\underline{v},t)\ ]$$

might yield some idea of the sensitivity of the problem to individual c_i. This information could then be used to devise a scheme for finding the optimal values $c_1^*,\ldots,c_{q+1}^*$. In general, however, a direct search technique must be employed. The method used here can be described as follows: let c_i^j, i=1,...,q+1, be the j^{th} estimate of the constants c_i and let

$\bar{J}^j$ be the resultant objective value. A better set of constants is found by putting

$$c_i^{j+1} = c_i^j + \varepsilon^j (M_i - z_i^+(t_p)), \quad i=1,\ldots,q, \qquad 5.38a$$

$$c_{q+1}^{j+1} = c_{q+1}^j + \varepsilon^j (J^j - c_{q+1}^j), \qquad 5.38b$$

and adjusting the value of ε^j to ensure $\bar{J}^{j+1} < \bar{J}^j$. Any other search technique not requiring information about derivatives could be used. Fortunately the evaluation of $\underline{z}^+(t_p)$ for a given set of constants c_i can be carried out by numerical quadrature. So that whatever search method is employed, the determination of c_i^*, $i=1,\ldots,q+1$, should not prove too time consuming.

5.5. An algorithm for periodic operation of nonlinear processes.

Having proposed a method for solution of problems linear in the state variables we shall now employ it to the solution of nonlinear problems. Let: k denote the k^{th} iteration; $\underline{u}^k(t)$, $0 \leqslant t \leqslant t_p$, be the k^{th} approximation to the optimal control; $\underline{x}^k(t)$, $0 \leqslant t \leqslant t_p$, be the periodic phase trajectory obtained by the repeated application of $\underline{u}^k(t)$ to system (5.1); $\underline{x}^{+k}(t_p)$ be the final value of $\underline{x}^+(t)$ obtained with $\underline{u}^k(t)$, $\underline{x}^k(t)$; and $\bar{J}^k = \psi(\underline{x}^{+k}(t_p))$ be the resultant value of the augmented objective.

The linearisation of systems (5.2) and (5.5) about the periodic trajectory $\underline{x}^k(t)$, $0 \leqslant t \leqslant t_p$, then yields

$$\dot{\underline{z}} = \left(\frac{\partial \underline{f}}{\partial \underline{x}}\right)^k \underline{z} + \left[\underline{f}(\underline{x}^k(t),\underline{v}) - \left(\frac{\partial \underline{f}}{\partial \underline{x}}\right)^k \underline{x}^k(t) \right],$$

$$\dot{\underline{z}}^+ = \left(\frac{\partial \underline{f}^+}{\partial \underline{x}}\right)^k \underline{z} + \left[\underline{f}^+(\underline{x}^k(t),\underline{v}) - \left(\frac{\partial \underline{f}^+}{\partial \underline{x}}\right)^k \underline{x}^k(t) \right], \qquad 5.39$$

where the superscript k indicates evaluation with $\underline{u}^k(t)$ and $\underline{x}^k(t)$. Now, Eqs.(5.39) and (5.10) are identical in form. The optimal control vector, $\underline{v}^*(t)$, $0 \leqslant t \leqslant t_p$, for the linearised system (5.39) could therefore be found by the method set out in the previous section if we make the following substitutions:

$$F(t) = \left(\frac{\partial \underline{f}}{\partial \underline{x}}\right)^k, \quad F^+(t) = \left(\frac{\partial \underline{f}^+}{\partial \underline{x}}\right)^k,$$

$$\underline{g}(\underline{v},t) = \underline{f}(\underline{x}^k(t),\underline{v}) - \left(\frac{\partial \underline{f}}{\partial \underline{x}}\right)^k \underline{x}^k(t), \qquad 5.40$$

$$\underline{g}^+(\underline{v},t) = \underline{f}^+(\underline{x}^k(t),\underline{v}) - \left(\frac{\partial \underline{f}^+}{\partial \underline{x}}\right)^k \underline{x}^k(t).$$

The optimal periodic end state for the linearised system (5.39) would then be given by

$$\underline{z}^{*k}(0) = \underline{z}^{*k}(t_p) = [I_n - \phi_1(0)]^{-1} \int_0^{t_p} \phi_1(t)\underline{g}(\underline{v}^{*k}(t),t)\, dt. \qquad 5.41$$

Now, if the linearised system was a true representation of the nonlinear system the optimal control for the latter would also be given by $\underline{u}^*(t)=\underline{v}^{*k}(t)$ and the periodic end state would be $\underline{x}^*(0)=\underline{x}^*(t_p)=\underline{z}^{*k}(0)$. Initially, however, $\underline{u}^k(t)$ and $\underline{v}^{*k}(t)$ are not close to each other, so that the linearised system is far from being a valid representation of the original nonlinear system.

We therefore choose a control vector $\underline{u}^{k+1}(t)$ which brings the two systems closer to each other. This can be done by putting

$$\underline{u}^{k+1}(t) = (1-\alpha^k)\,\underline{u}^k(t) + \alpha^k\,\underline{v}^{*k}(t),$$

$$\underline{x}^{k+1}(0) = (1-\alpha^k)\,\underline{x}^k(0) + \alpha^k\,\underline{z}^{*k}(0), \qquad 5.42$$

where α^k is a positive scalar, $0 \ll \alpha^k \ll 1$. Now, if α^k is sufficiently small, we can find a periodic phase trajectory, $\underline{x}^{k+1}(t)$, such that $\bar{J}^{k+1} < \bar{J}^k$. The process could then be linearised about $\underline{x}^{k+1}(t)$, $0 \ll t \ll t_p$, and the resulting linear problem solved and the whole procedure could then be repeated. In this way $\underline{u}^k(t)$ could be made closer and closer to $\underline{v}^{*k}(t)$, and the linearised system gradually becomes a better approximation of the nonlinear system. Then, as the iterations proceed, the value of α^k could be increased until no further reduction in $\bar{J}$ can be obtained with $\alpha^k = 1$. In this case the linearised system has become a valid representation of the original nonlinear system and the optimal control is given by $\underline{u}^*(t)= \underline{v}^{*k}(t)$.

To summerise, the solution of the nonlinear problem involves the following steps:

Step 1. Guess a period t_p; a nominal periodic control, $\underline{u}^0(t)$, $0 \leqslant t \leqslant t_p$; an initial state $\underline{x}^0(0)$; and a small positive scalar α^0.

Step 2. Integrate the nonlinear system (5.5) forward with $\underline{u}^k(t)$ and $\underline{x}^k(0)$ until periodicity is established. Store the resultant periodic phase trajectory $\underline{x}^k(t)$, $0 \leqslant t \leqslant t_p$.

Step 3. Calculate $\underline{x}^{+k}(t_p)$ from

$$\underline{x}^{+k}(t_p) = \int_0^{t_p} \underline{f}^+(\underline{x}^k(t), \underline{u}^k(t))\, dt,$$

and determine $\bar{J}^k = \Psi(\underline{x}^{+k}(t_p))$.

Step 4. With $F(t)$, $F^+(t)$, $\underline{g}(\underline{v},t)$, and $\underline{g}^+(\underline{v},t)$ as defined by Eqs.(5.40) solve the lincarised problem. Store the resulting $\underline{v}^{*k}(t)$ and find $\underline{z}^{*k}(0)$ from Eq.(5.41).

Step 5. Determine $\underline{u}^{k+1}(t)$ and $\underline{x}^{k+1}(0)$ from Eqs.(5.42) and repeat Steps 2 and 3 with $\underline{u}^{k+1}(t)$. Reducing α^k, if necessary, to ensure that $\bar{J}^{k+1} < \bar{J}^k$.

Step 6. Put k=k+1 and repeat Steps 4 to 6, gradually increasing the value of α^k, until no further reduction in $\bar{J}^k$ can be obtained with $\alpha^k = 1$.

Using the above procedure the solution to the nonlinear problem can be obtained by the successive solution of the linearised problem (Step 4). The scalar α^k is in effect the gain of the procedure: if it is chosen too large or increased too rapidly divergence may occur. In our version of the algorithm its value is halved whenever $\bar{J}^{k+1} > \bar{J}^k$, and it is tentatively doubled if $\bar{J}^{k+1} \leqslant \bar{J}^k$ for two successive iterations. The only numerical integration involved (Step 2) is in order to find a periodic trajectory corresponding to a given periodic control. This might prove time consuming if the integration has to be carried out over a large number of periods. However, after the first iteration, the periodic end states and the control for successive iterations will be close to each other; so that the integration should extend over a decreasing number of periods. An alternative method is to use a Newton-Raphson iteration technique to match the boundary conditions at the begining, $\underline{x}(0)$, and the end, $\underline{x}(t_p)$, of a single period as suggested by Horn and Lin [47].

Finally, to use the above procedures with the usual integral objective functions we need only to modify Eq.(5.37) to

$$\bar{J} = \psi(\underline{x}^+(t_p)) = x^+_{q+1}(t_p) + \frac{\omega}{2} \sum_{i=1}^{q} (x^+_i(t_p) - M_i)^2 . \quad 5.43$$

The constant c^*_{q+1} may now be arbitrarily set to - 1, and the search procedure on c_i is confined to the first q components $c_1, \ldots, c_q$.

5.6. Computational results

The problems arising in the unsteady operation of a continuous process can be divided into three types: those with

(a) an ordinary integral objective function and no integral side constraints,

(b) an ordinary integral objective function and integral side constraints,

(c) a ratio-integral objective function and integral side constraints.

The results presented below reflect the application of the proposed algorithm to the determination of optimal periodic operation of a continuous process for each of the above cases. All the computational results were obtained on an I.B.M. 360 computer employing the single computer program fully described and presented in Appendix 5. All the numerical calculations were performed in double precision arithmatic.

The process examined is one used throughout this thesis; namely the isothermal operation of a continuous stirred tank reactor with the consecutive-competing reaction scheme

$$S_1 + S_2 \longrightarrow S_3,$$

$$S_2 + S_3 \longrightarrow S_4.$$

In keeping with previous chapters it is assumed that the inlet concentrations in dynamic operation cannot exceed their corresponding steady values and the flow rate remains fixed throughout each operation. The process is then described by

$$\dot{x}_1 = wu_1 - wx_1 - \alpha_1 x_1 x_2,$$

$$\dot{x}_2 = wu_2 - wx_1 - \alpha_1 x_1 x_2 - \alpha_2 x_2 x_3,$$

$$\dot{x}_3 = - wx_3 + \alpha_1 x_1 x_2 - \alpha_2 x_2 x_3,$$

$$\dot{x}_4 = - wx_4 + \alpha_2 x_2 x_3,$$

where

$$x_i = A_i/A_{1fs},\ i=1,..,4;\ u_i = A_{if}/A_{1fs},\ i=1,2;$$
$$w = F/F_s;\ \theta = tF_s/V;\ \alpha_i = k_i V A_{1fs}/F_s,\ i=1,2;$$

and the inputs u_1 and u_2 must satisfy the following constraints

$$0 \leqslant u_1(\theta) \leqslant 1, \text{ for any } \theta,$$

$$u_2(\theta) = \frac{A_{2fs}}{A_{1fs}} u_1(\theta), \text{ for any } \theta$$

5.6.1. Case (a): an ordinary integral objective with no side constraints.

Consider the problem where for a given flow rate, w, the objective is to maximise the difference in the output of the

desired product, S_3, and the undesired one, S_4. This is identical to the problem examined in chapter 3 and the objective to be minimised is given by

$$J = \int_0^{\theta_p} w\ (x_4(\theta) - x_3(\theta))\ d\theta.$$

For this case, there are no integral side constraints, q=0, the only unknown constant is c_1 which may be taken as - 1. The augmented objective $\bar{J}$ (see Eq.5.43) is therefore given by

$$\bar{J} = \psi(\underline{x}^+(\theta_p)) = x_1^+(\theta_p)$$

where

$$x_1^+(\theta_p) = \int_0^{\theta_p} w\ (x_4(\theta) - x_3(\theta))\ d\theta.$$

The problem was solved for various rate constant ratios and a range of flow rates. As in chapter 3, irrespective of the starting policies, for flow rates above a critical value, $w > w_c$, a final steady policy at the maximum allowable level, and for flow rates below the critical value, $w < w_c$, a final on-off policy were obtained.

Results from a typical run, with θ_p=20 and $w=0.25 < w_c$, are represented in figures 5.1a and 5.1b where the control policy for the nonlinear system, $\underline{u}^k(\theta)$, and the corresponding optimal control for the linearised system, $v^{*k}(\theta)$, are shown

for every fifth iteration. The results for the last six iterations are shown seperately in figures 5.2a and 5.2b. The objective function, $\bar{J}$, and the gain of the procedure, α, for each iteration are also shown in figures 5.3a and 5.3b. Identical results were obtained when the discretisation interval, $\Delta\theta$, was first halved and then quartered.

The control, $u^k(\theta)$, changes very rapidly for the first twenty iterations. It is then modified very slightly between iterations 20 and 45 before rapidly converging to its final on-off form over the last six iterations. To understand the behaviour of the algorithm it is sufficient to recall that the linearised system is a valid representation of the nonlinear system only if $u^k(\theta)$ and the resulting $v^{*k}(\theta)$ are "close" to each other.

Initially the control $u^0(\theta)$ is far from being optimal, we can therefore make large changes without the linearisation holding true. This is reminiscent of the large initial steps possible in a first order gradient search method when the initial guess is not near the optimum. The objective, $\bar{J}$, is therefore reduced sharply for the first eight iterations and the gain, α, builds up rapidly.

However, as the final value of $\bar{J}$ is approached the procedure becomes more sensitive to the validity of the linearisation and the changes in successive $u^k(\theta)$ become smaller. The analogy is with the reduced steps in a first order gradient search method as the optimum is approached. Consequently,

between iterations 20 and 45 the algorithm concentrates on bringing $v^{*k}(\theta)$ and $u^{k}(\theta)$ closer to each other and so make the linearisation more valid. This is reflected in a flat plateau in the objective, $\bar{J}$, and a reduction followed by oscillation in the gain, α.

However, once $u^{k}(\theta)$ and $v^{*k}(\theta)$ are made sufficiently close to each other for the linearisation to be valid, the gain of the procedure can be increased and the final policy is approached rapidly with a final dip in the value of the objective, $\bar{J}$. This final acceleration does not have an analogy with a first order gradient method. In fact it resembles the behaviour of second order gradient methods which require a good initial guess but converge rapidly near the optimum.

To test the algorithm further a run was made with a period θ_p=40, while keeping all other parameters and the initial policy the same. Identical results were obtained and figure 5.4 is a coarse representation of the successive policies obtained. It is interesting to note that doubling the period has had no effect, the results obtained at each step being exactly double those for a period of θ_p=20. This validates the point that in continuous periodic operations only a single period need be considered.

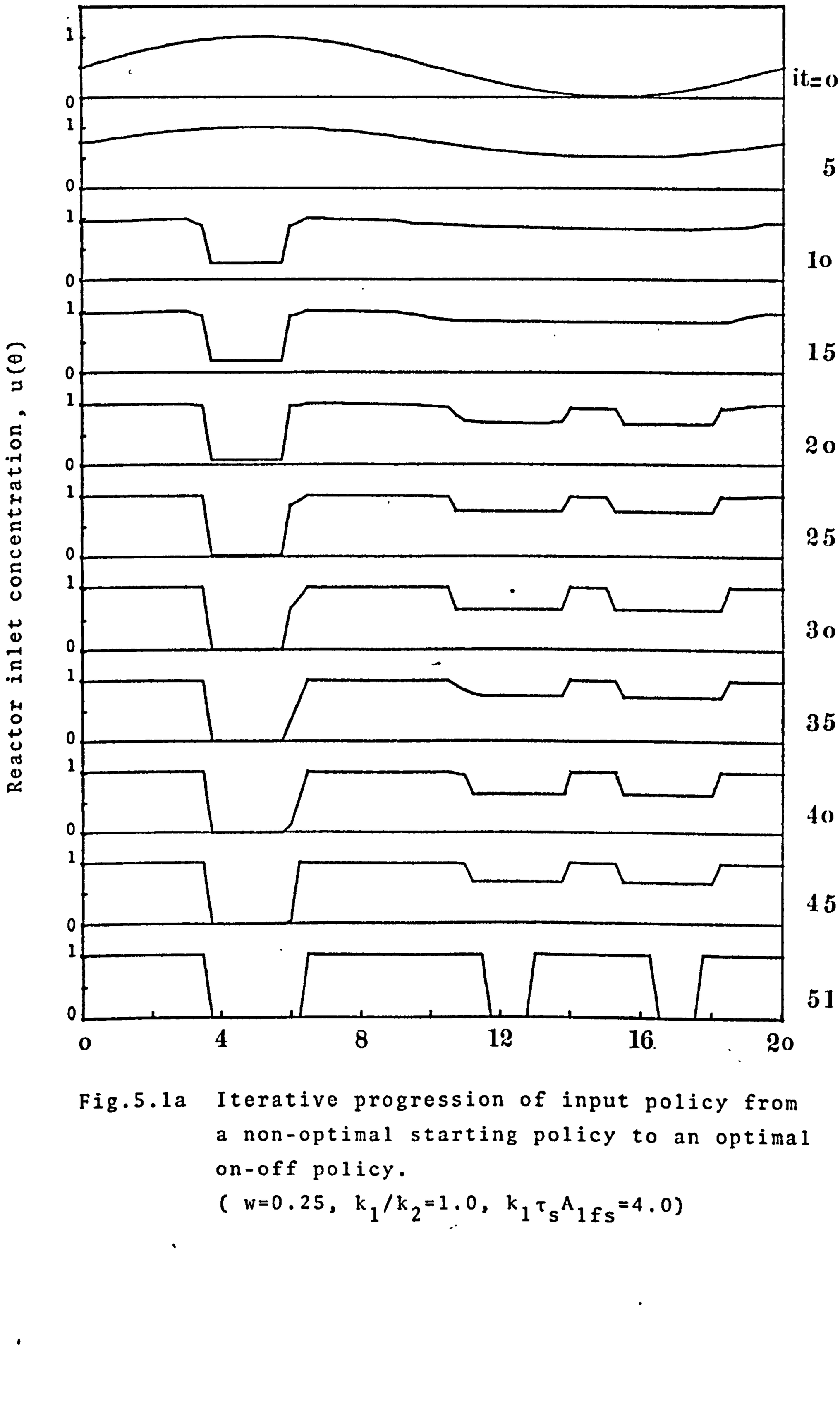


Fig.5.1a Iterative progression of input policy from a non-optimal starting policy to an optimal on-off policy.
($w=0.25$, $k_1/k_2=1.0$, $k_1\tau_s A_{1fs}=4.0$)

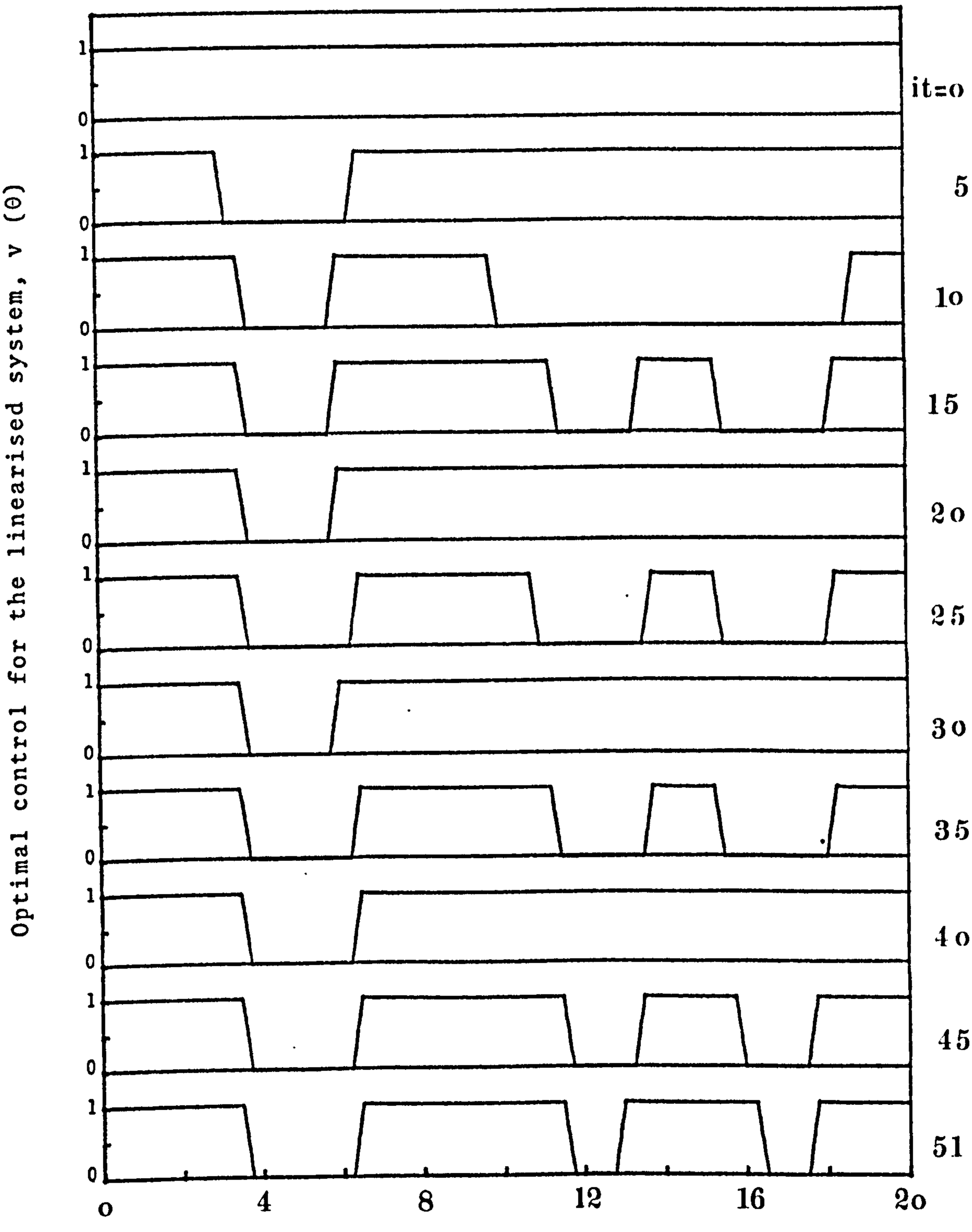


Fig.5.1b Successive optimal controls for the linearised system.
(all parameters as in Fig.5.1a)

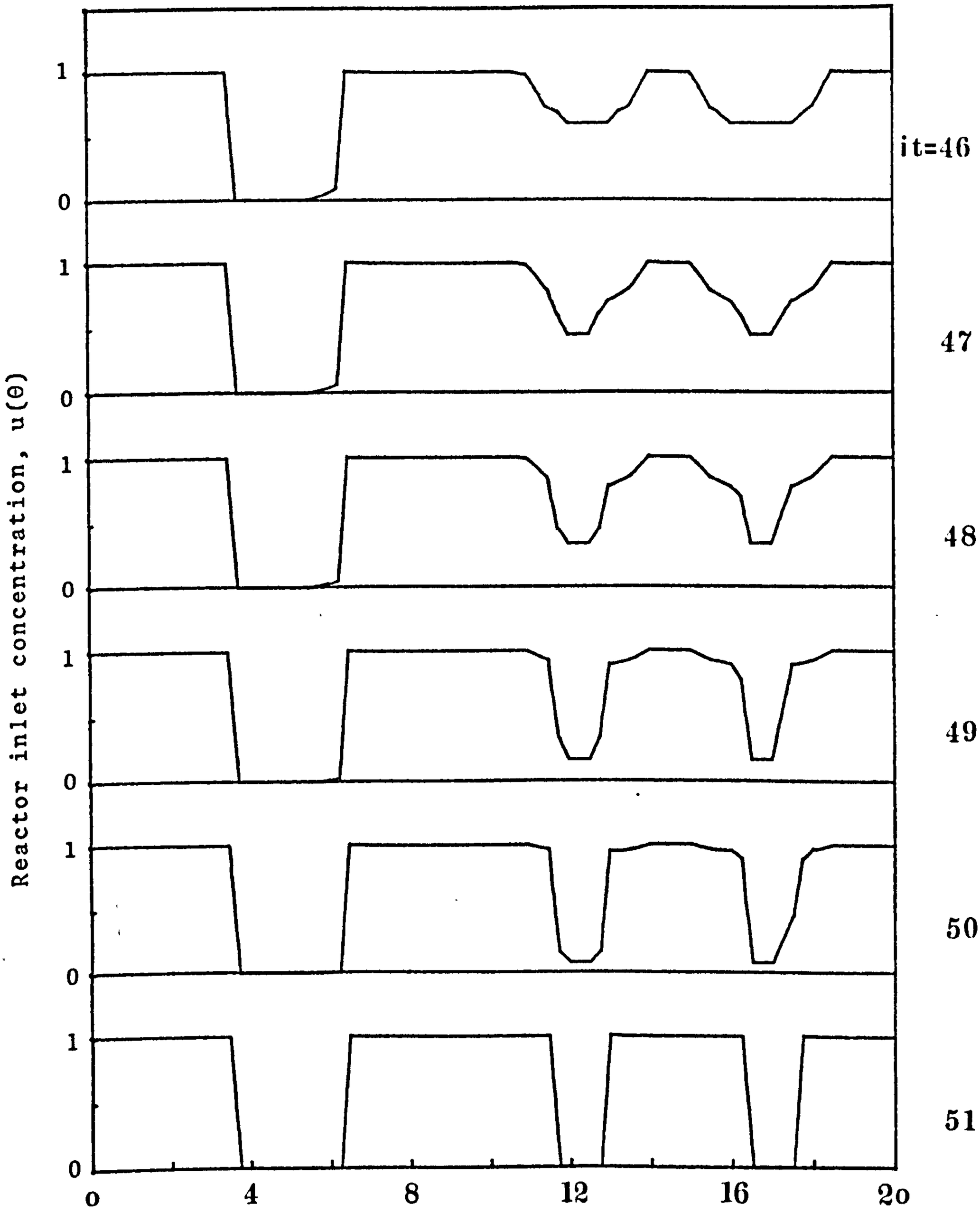


Fig.5.2a Iterative progression of input policy from a non-optimal policy to an optimal on-off policy.
(all parameters as in Fig.5.1a)

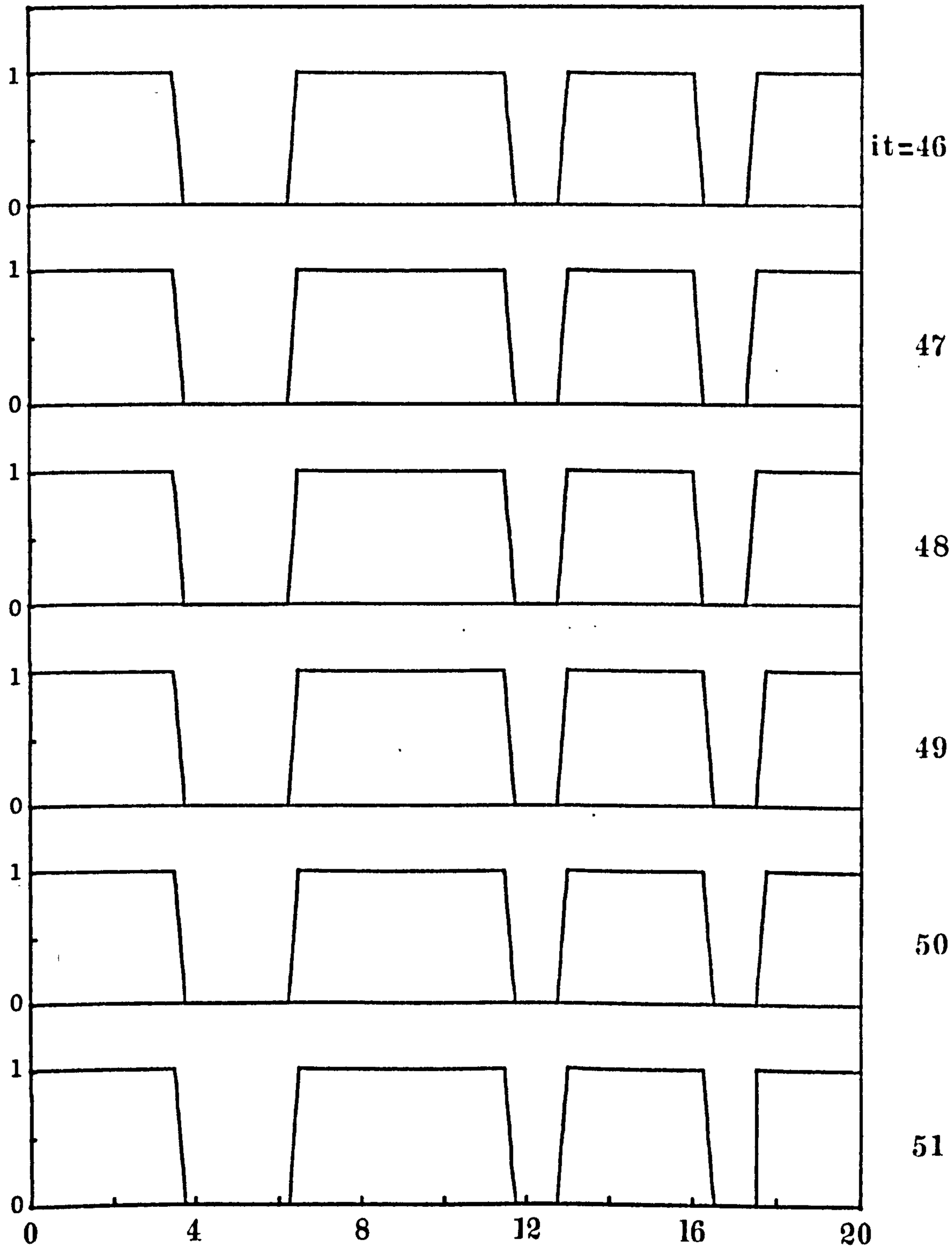


Fig.5.2b Successive optimal controls for the linearised system.
(all parameters as in Fig.5.1a)

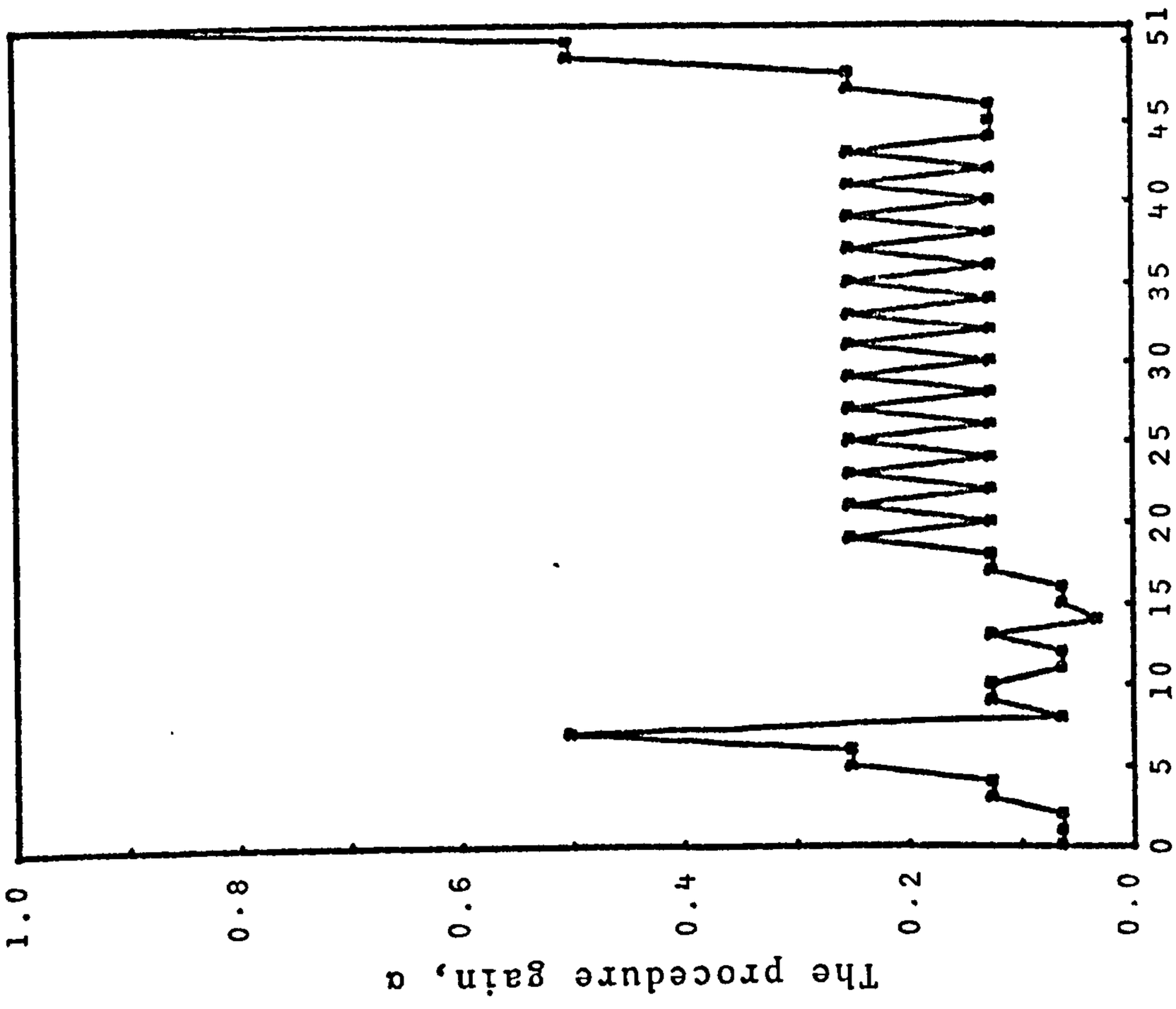


Fig.5.3b The procedure gain, α, at each iteration.
(all parameters as in Fig.5.1a)

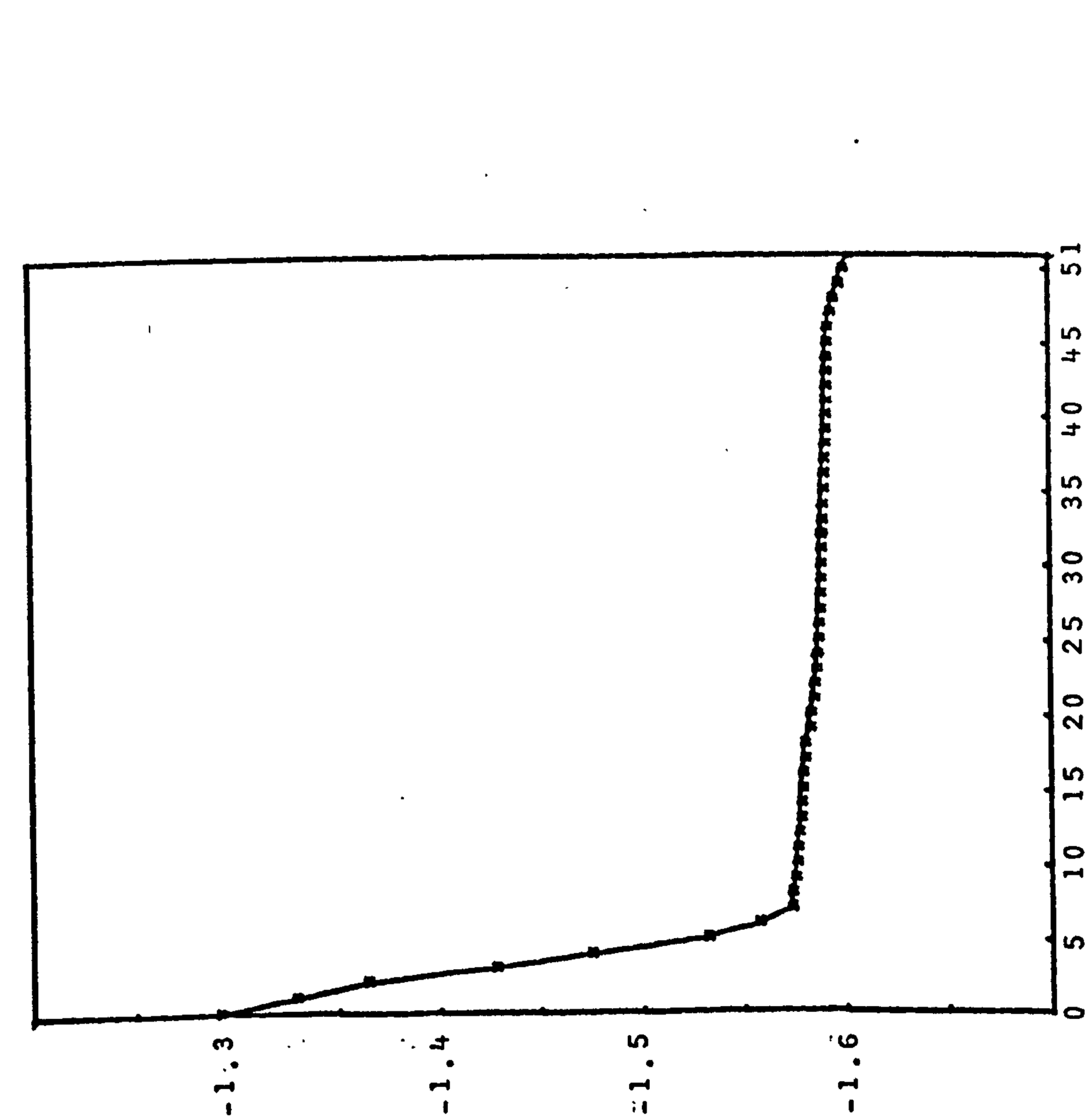


Fig.5.3a The objective function at each iteration.
(all parameters as in Fig.5.1a)

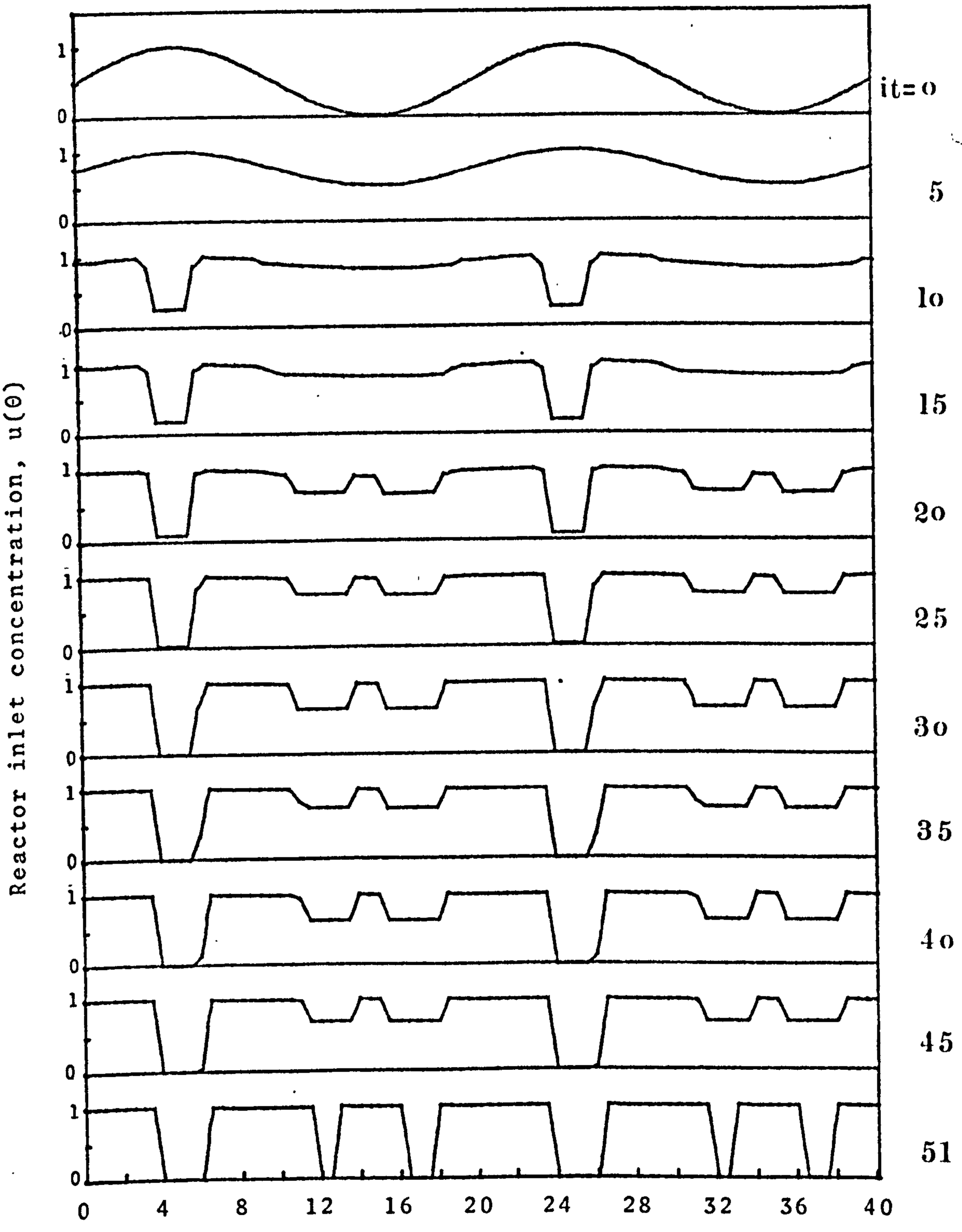


Fig.5.4. Iterative progression of input policy from a non-optimal starting policy to an optimal on-off policy.
(all parameters as in Fig.5.1a)

5.6.2. Case (b): an ordinary integral objective function with integral side constraint

Consider the problem of maximising the output of the desired product, S_3, while a given amount, M, of the reactant is to be fed to the reactor over each period, θ_p. Now, under optimal steady conditions (w=1 and u(θ)=1) the amount of the reactant reaching the reactor over the period, θ_p, is given by

$$\int_0^{\theta_p} w\, u(\theta)\, d\theta = \int_0^{\theta_p} 1x1\, d\theta = \theta_p \quad \text{mass units.}$$

So that if the amount of reactant available, M, is taken equal to that in the steady operation, $M=\theta_p$, the problem becomes identical to that examined empirically in chapter 2.

The objective function to be minimised in this case is given by

$$\int_0^{\theta_p} - w\, x_3(\theta)\, d\theta,$$

and the integral side constraint takes the form

$$\int_0^{\theta_p} w\, u(\theta)\, d\theta = M = \theta_p \text{ mass units.}$$

There are two unknown constants, c_1, c_2, involved in this case; c_2^* may be arbitrarily taken as -1, and a search is necessary to determine the optimal value of c_1 at each iteration. The augmented objective, $\bar{J}$, takes the form (see Eq.5.43):

$$\bar{J} = (\underline{x}^{+}(\theta_p)) = x_2^{+}(\theta_p) + \frac{\omega}{2} (x_1^{+}(\theta_p) - \theta_p)^2$$

where

$$x_1^{+}(\theta_p) = \int_0^{\theta_p} w\, u(\theta)\, d\theta \quad \text{and} \quad x_2^{+}(\theta_p) = \int_0^{\theta_p} - w\, x_3(\theta)\, d\theta .$$

It should be pointed out that if the weighting scalar, ω, is chosen too small, the first term in, $\bar{J}$, becomes dominant and thusthe side constraint cannot be accurately satisfied. On the other hand, if ω is chosen to large the second term will be dominant and the algorithm tends to fulfill the side constraint without minimising, $\bar{J}$. This was confirmed by some initial computation; two runs made with ($w=2.0$, $k_1/k_2=10.0$, $\theta_p=2.0$) identical parameters and different values of ω gave the following results:

ω	$(x_1^{+}(\theta_p)-\theta_p)^2$	$x_2^{+}(\theta_p)$	$\bar{J}$
1	0.38242×10^{-1}	-o.654474	-0.635353
50	0.49303×10^{-31}	-0.598430	-0.598430

For this problem the objective,which is the production rate of the desired product, S_3, increases as more material is fed to the reactor. Consequently, if ω is chosen too small the amount reaching the reactor is higher than the set value(i.e. the first term in $\bar{J}$ is positive rather than zero). The objective obtained however is smaller than the case where the set amount of reactant is fed to the reactor(i.e. the second term in $\bar{J}$ is more negative).

In other words, the increased production rate obtained at the expense of violating the side constraint dominates the augmented objective. Naturally, to decide between the two modes of operation one must use real economic data. In the absence of such data, we shall require that the side constraint be fully satisfied. A suitable range for ω was then found to be $10 \leqslant \omega \leqslant 50$.

The problem was then solved for various rate constant ratios and flow rates ranging between 1.25 to 5 times the steady flow rate. In all cases where ω was chosen properly, the procedure rapidly converged to a final on-off policy. Figures 5.5a and 5.5b show the successive control policies for the nonlinear system, $u^k(\theta)$, and the corresponding optimal controls for the linearised system, $v^*(\theta)$, for a typical run. The objective function, $\bar{J}$, and the gain of the procedure, α, are presented in Figures 5.6a and 5.6b.

In all cases tested the optimal periodic end states for the nonlinear system, $\underline{x}^*(0)=\underline{x}^*(\theta_p)$, and the linearised system, $\underline{z}^*(0)=\underline{z}^*(\theta_p)$ differed by less than one percent. The optimal value of the objective function for the two systems where however much closer. Some typical results are presented below:

θ_p	$\bar{J}^*$(linear)	$\bar{J}^*$(nonlinear)	$(\underline{z}^*(0)-\underline{x}^*(0))^2$
1.25	-.605462	-.605415	1.95×10^{-6}
1.50	-.603989	-.603856	2.50×10^{-6}
1.75	-.601807	-.601659	4.82×10^{-6}
2.00	-.599389	-.599350	4.08×10^{-6}
2.25	-.601250	-.600610	5.43×10^{-6}

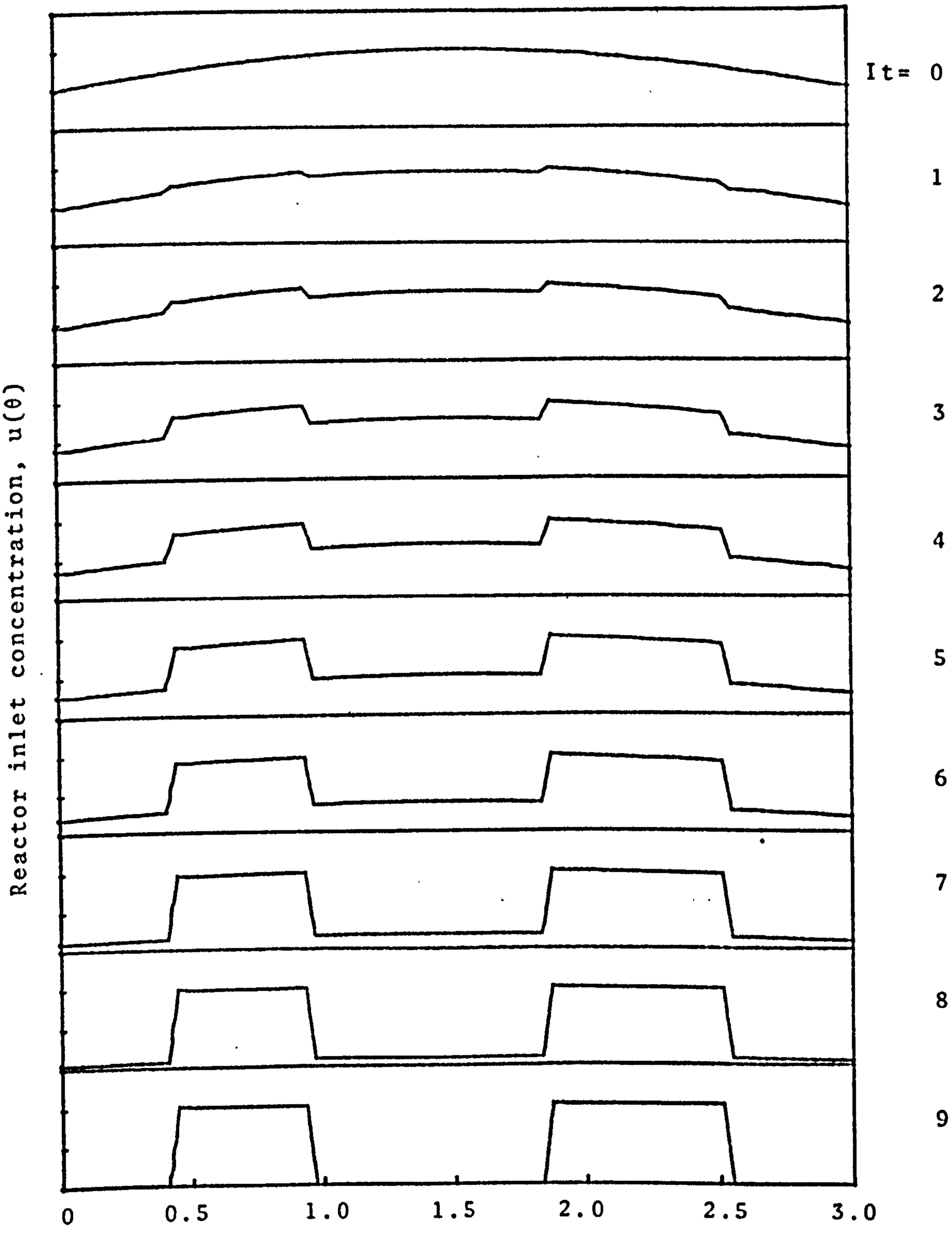


Fig.5.5a Iterative progression of input policy from a non-optimal starting policy to an on-off policy.

($w=2.5$. $k_1/k_2=10.0$, $k_1\tau_s A_{1fs}=54.785$)

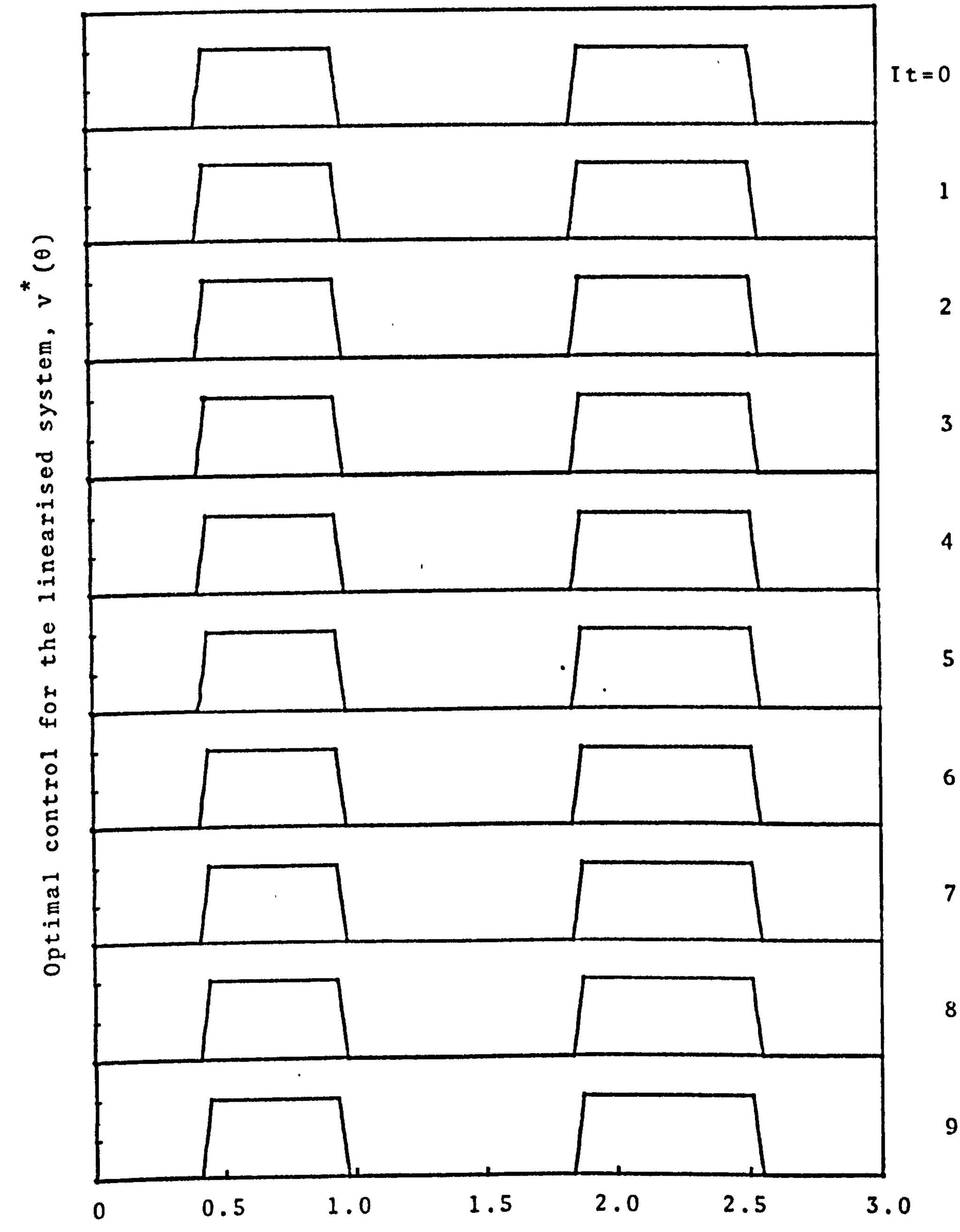


Fig.5.5b Successive optimal controls for the linearised system.
(all parameters as in Fig.5.1a)

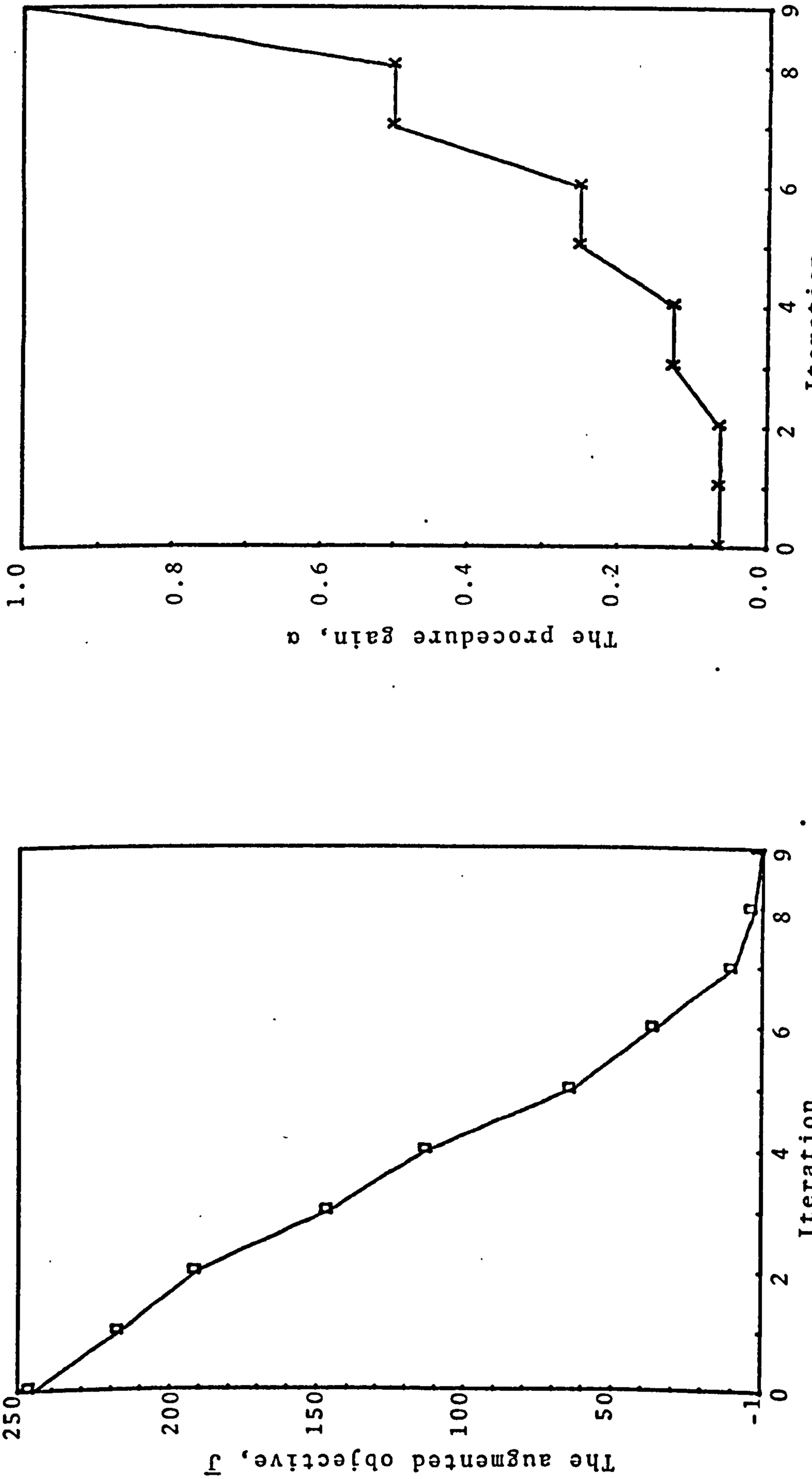


Fig.5.6b The procedure gain, α, at each iteration.
(all parameters as in Fig.5.5a)

- Fig.5.6a The augmented objective function at each iteration.
(all parameters as in Fig.5.5a)

5.6.3. Case (c): a ratio-integral objective function with integral side constraint

Consider the problem of maximising the selectivity of the desired product, S_3, while a given amount, M, of the reactant is to be fed to the reactor over each period, θ_p. As for the previous case if M is taken as $M=\theta_p$, the problem becomes identical to that examined empirically in chapter 2.

The objective function to be minimised in this case is given by

$$\frac{\int_0^{\theta_p} - w\, x_3(\theta)\, d\theta}{\int_0^{\theta_p} w\, (\, u(\theta) - x_1(\theta)\,)\, d\theta},$$

and the integral side constraints once again has the form

$$\int_0^{\theta_p} w\, u(\theta)\, d\theta = M = \theta_p \quad \text{mass units}$$

There are three unknown constants, c_1, c_2, c_3, involved in this case; c_3^* may be arbitraily set to -1, and a search is necessary to determine the optimal values of c_1 and c_2 at each iteration. The augmented objective function now takes the form (see Eq.5.37)

$$\bar{J} = \frac{1}{2}\left(c_2 - \frac{x_3^+(\theta_p)}{x_2^+(\theta_p)} \right)^2 - \frac{\omega}{2}\left(x_1^+(\theta_p) - \theta_p \right)^2$$

where $x_1^+(\theta_p)$ and $x_2^+(\theta_p)$ are as in Case (b) and $x_3^+(\theta_p)$ is given by

$$x_3^+(\theta_p) = \int_0^{\theta_p} w \ (\ u(\theta) - x_1(\theta) \ ) \ d\theta.$$

As in the previous case, the results obtained were sensitive to the value of the scalar, ω. In this case, however, the objective function, which is the selectivity of the desired product, S_3, is at its highest when a minimal amount of raw material is used. Consequently, if ω is chosen too small the amount of reactant fed to the reactor is less than the set value. However, the higher selectivity obtained by violating the constraint is at the expense of production rate. So that an economic appraisal is necessary to determine the best mode of operation.

The problem was solved for various rate constant ratios and a range of flow rates. In all cases where ω was chosen properly, the procedure rapidly converged to a final on-off policy. The results obtained showed the same characteristics as those of Case (b): the optimal value of the objective function for the nonlinear and the linearised systems were very close to each other and the optimal periodic end states differed by less than one percent.

The successive control policies for the nonlinear system, $u^k(\theta)$, and the corresponding optimal controls for the linearised system, $v^{*k}(\theta)$, for a run with θ_p=1.5, are shown in Figures 5.7a 5.7b. The phase trajectory for this run is presented in Figure 5.8. The double periodicity of the result for this case is immediately apparent.

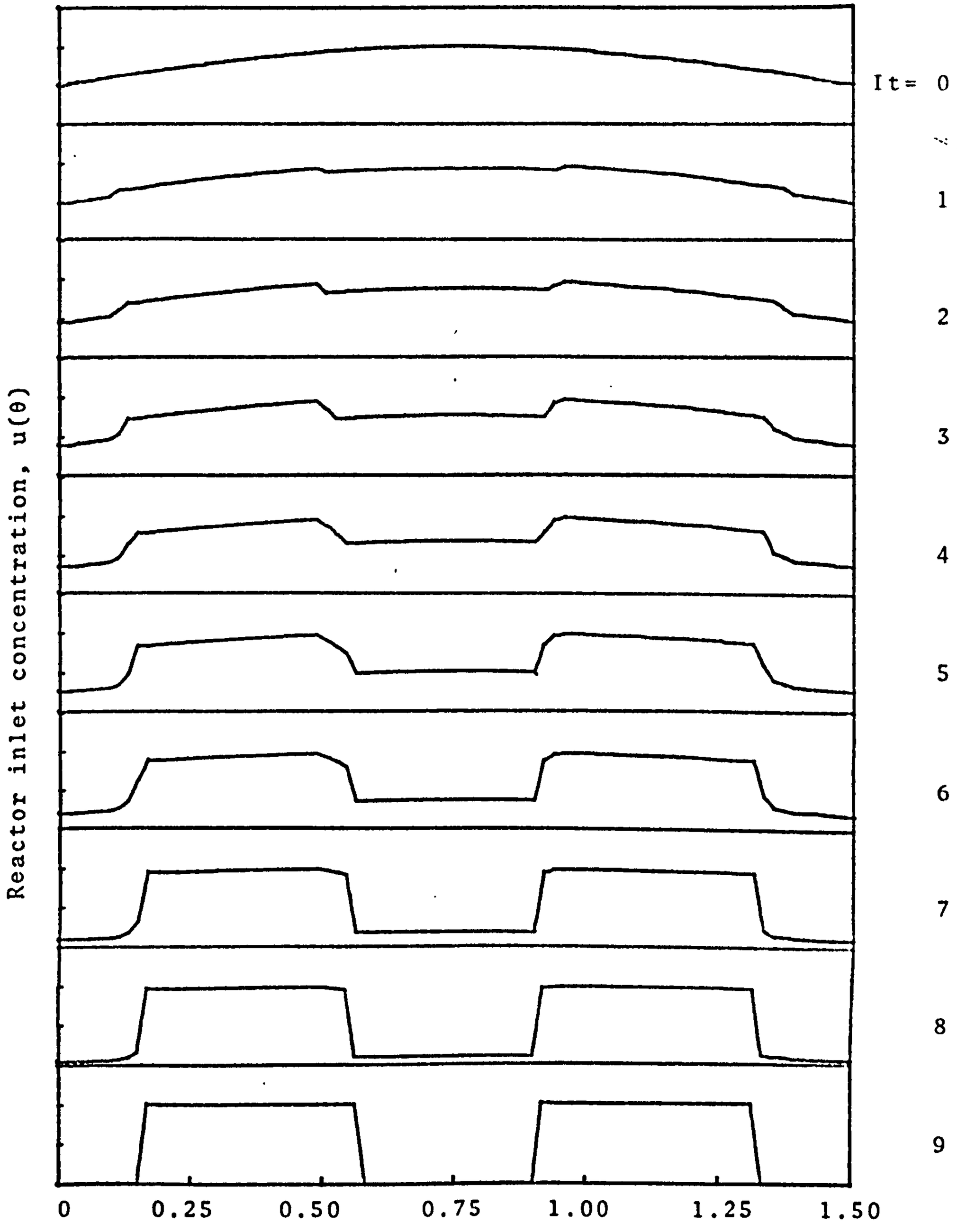


Fig. 5.7a Iterative progression of input policy from a non-optimal starting policy to an on-off policy.

($w=1.8181818$, $k_1/k_2=1$, $k_1\tau_s A_{1fs}=4.0$)

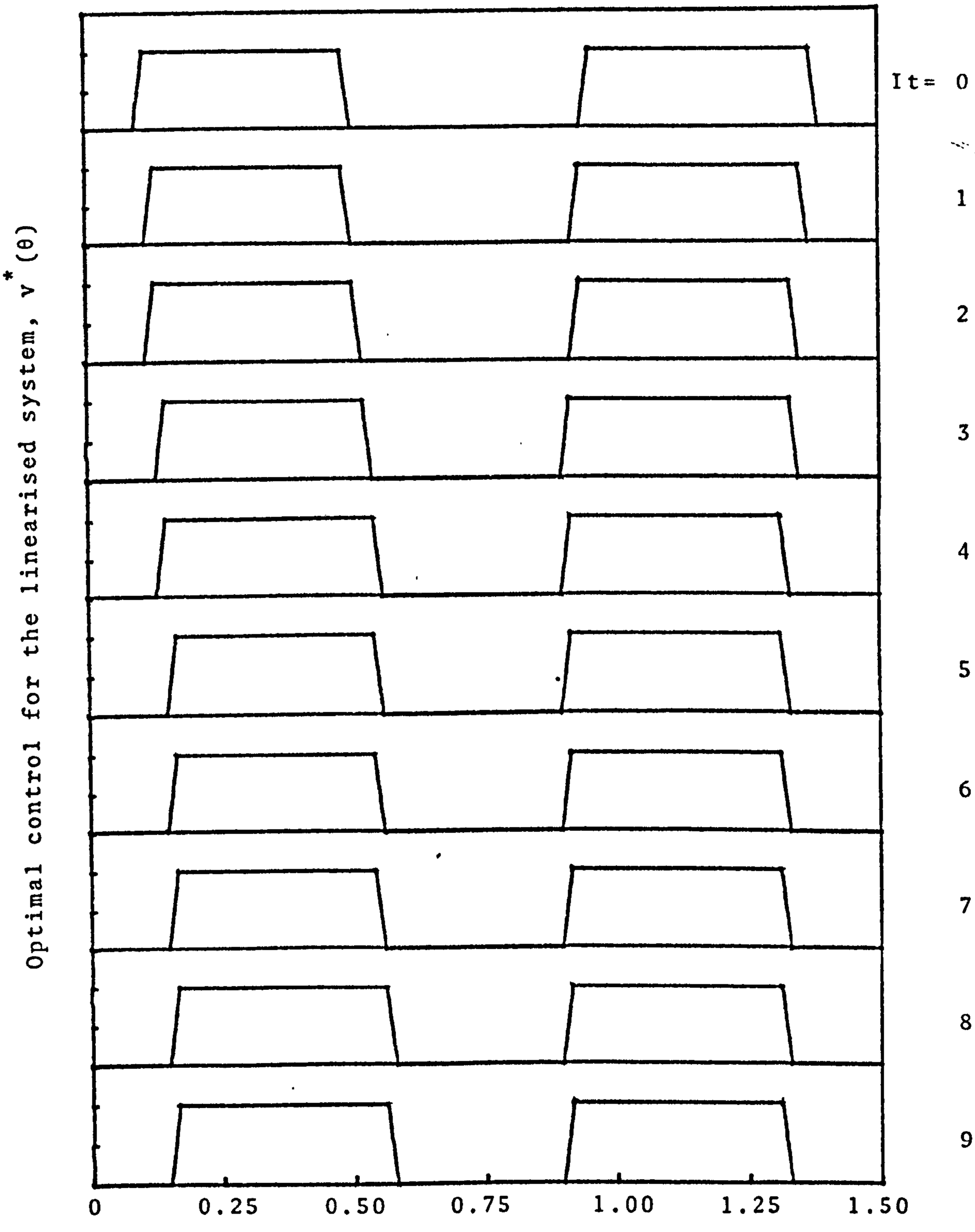


Fig.5.7b Successive optimal controls for the linearised system.

(all parameters as in Fig.5.7a)

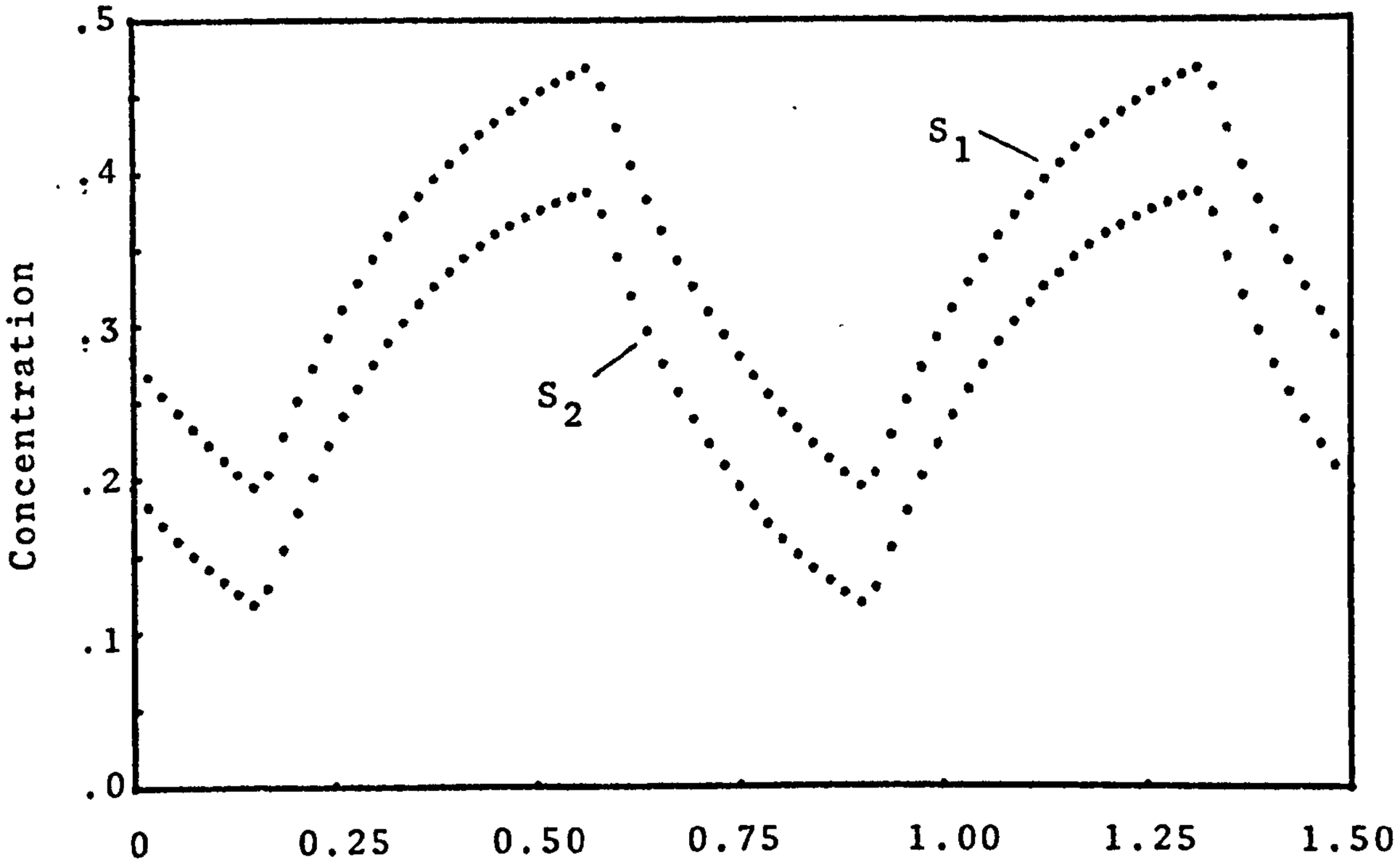


Fig.5.8a Periodic reactant concentrations

(all parameters as in Fig.5.7a)

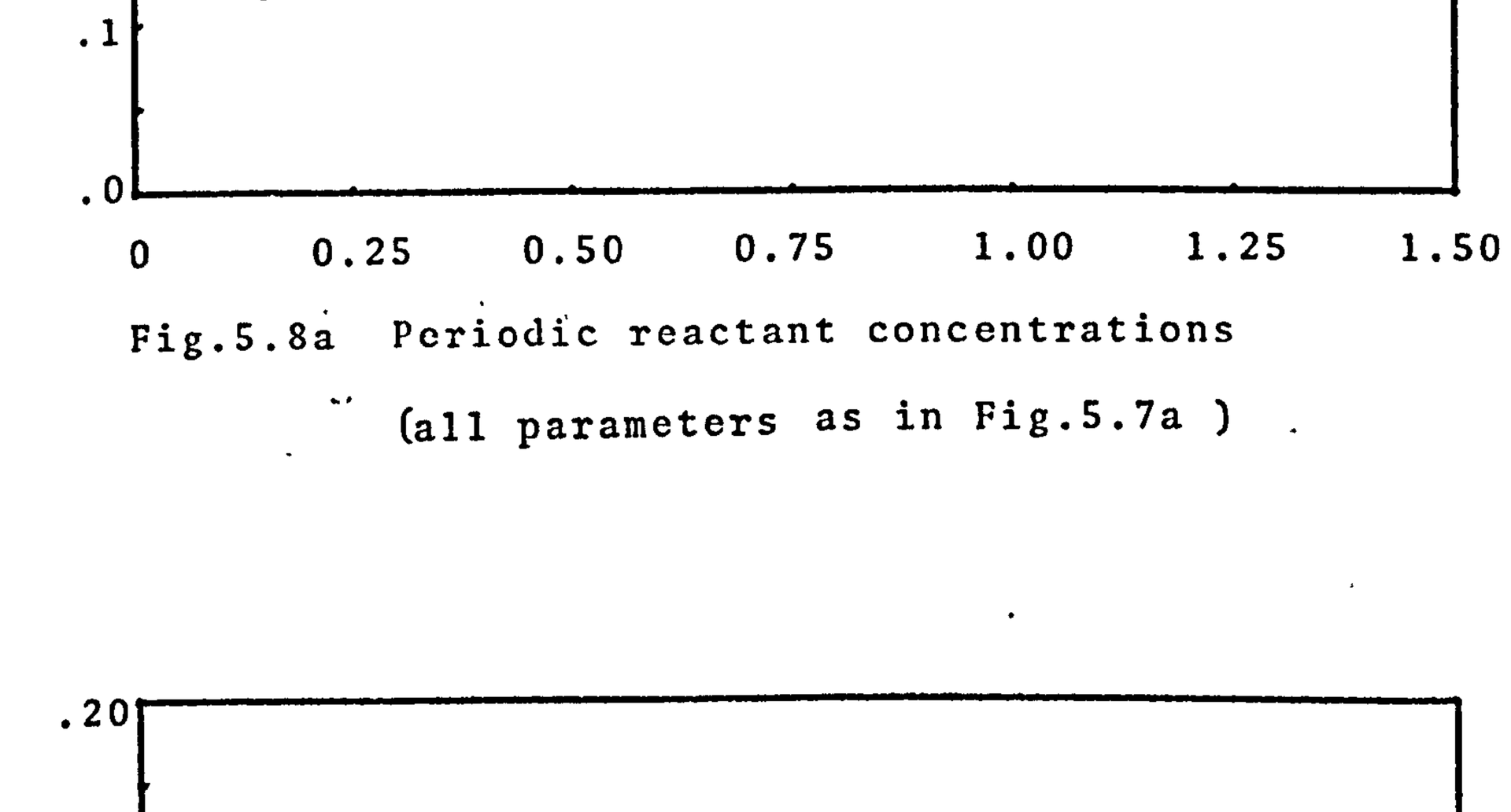


Fig.5.8b Periodic product concentrations

(all parameters as in Fig.5.7a)

5.6.4. Discussion

The proposed algorithm does not take into account the optimality of the period, θ_p. In other words, the final control policy obtained is the best periodic input profile within a given period, θ_p. Now, as we have already seen, doubling the period does not change the results obtained. This is because the algorithm which is specifically designed for periodic operation cannot distinguish between a periodic operation with a period θ_p or one with an exact multiple of θ_p. Consequently, unless the period chosen is an exact multiple of the optimal period, θ_p^*, better results may be obtained by gradually adjusting the period used.

The results obtained for Case (a) in chapter 3 indicated that the final optimal strategy was on-off periodic and unimodal. That is the optimal periodic waveform was composed of a train of identical unsymmetric pulses. A typical result is shown in Figure 3.3. The periodic policy found by the proposed method for identical parameters, an equal storage interval, and a period θ_p=20, is shown in Figure 5.1a. In contrast to chapter 3, the final on-off policy obtained is not unimodal in nature. The period, θ_p, was then gradually increased and the optimal control problem was solved again. The best results were obtained for θ_p=30 and are presented in Figures 5.9a and 5.9b. The final strategy obtained in this case is clearly unimodal and the waveform found is identical to that found in chapter 3.

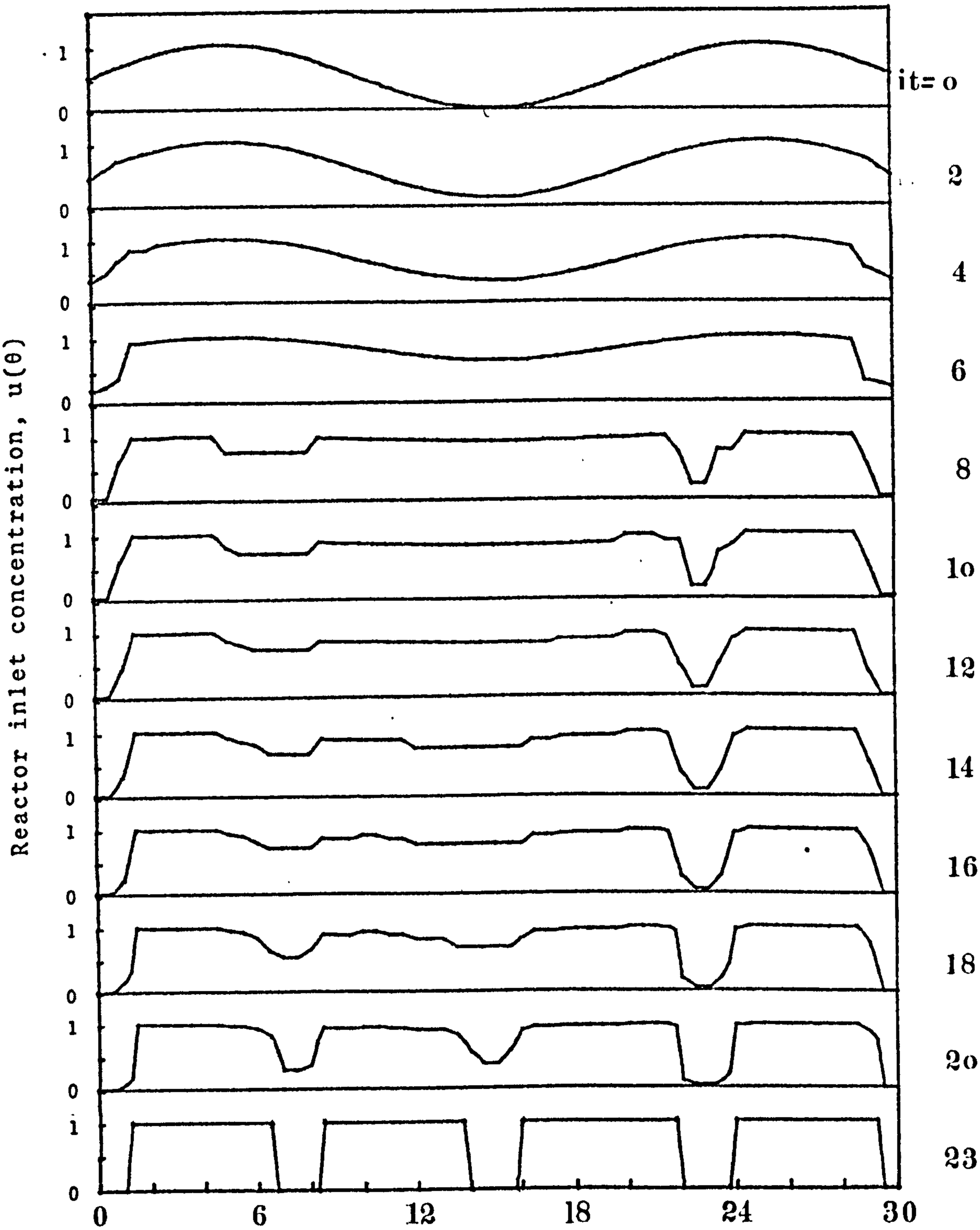


Fig.5.9a Iterative progression of input policy from a non-optimal starting policy to a unimodal on-off policy.

($w=0.25$, $k_1/k_2=1.0$, $k_1\tau_s A_{1fs}=4.0$)

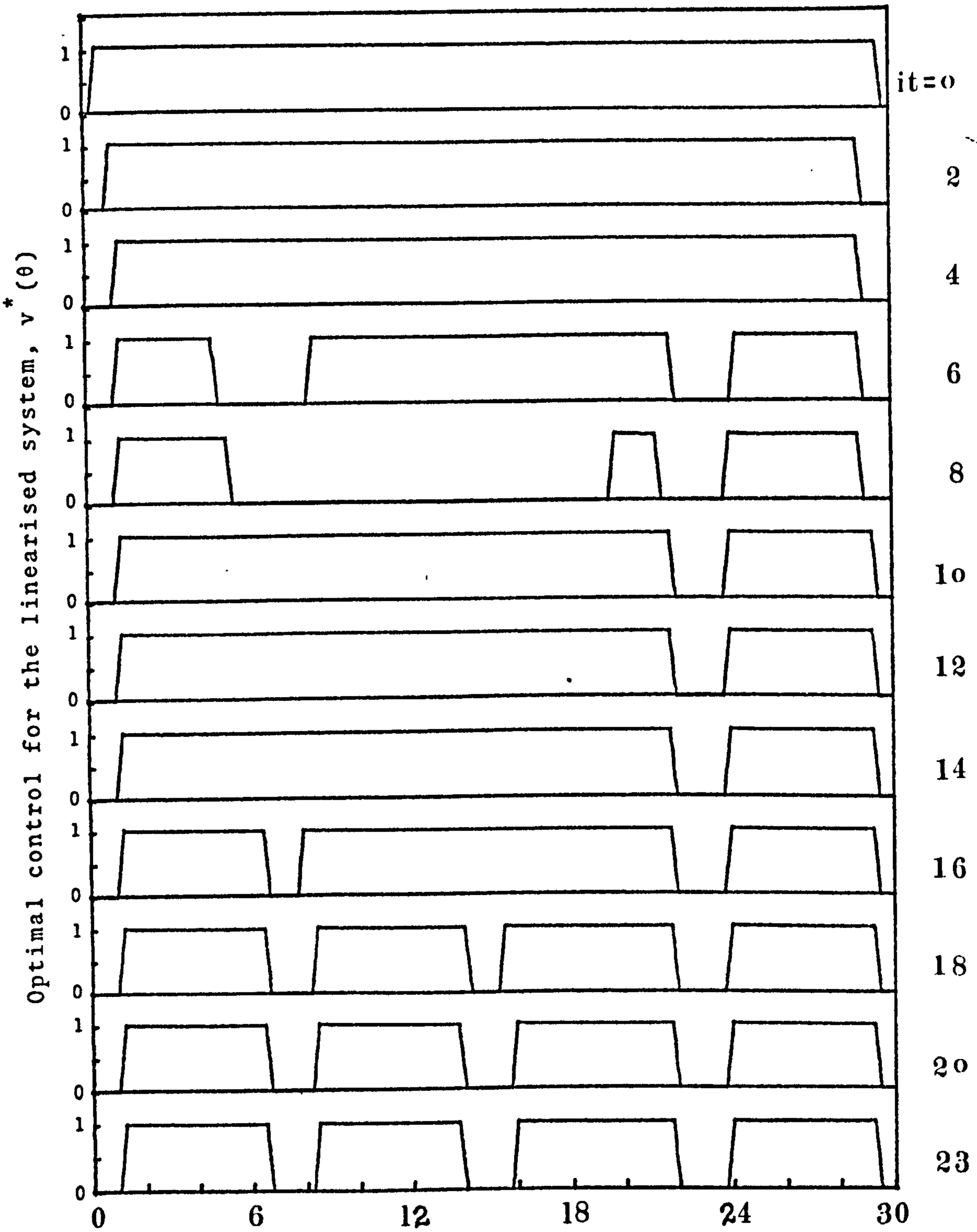


Fig.5.9b Successive optimal controls for the linearised system.
(all parameters as in Fig.5.9a)

The computational requirements for the method of chapter 3 and the proposed method for identical parameters and an equal discretisation interval are compared below:

	operating interval	objective J*	computation time (min)	array area (kilo bytes)
chapter 3	300	-0.160922	65	77
proposed algorithm	20	-0.160261	13	18
	40	-0.160261	20	36
	30	-0.160836	7	27

The enormous reductions in computation time and storage obtained by confining our attention to much shorter operating intervals are immediately obvious. It is interesting that doubling the period from 20 to 40 does not double the computation time used. More significantly, when the period chosen, θ_p=30, is an exact multiple of the optimal period the number of iterations and the computation time required are almost halved. The objective function, J, and the gain of the procedure, α, for this case are shown in figures 5.10a and 5.10b. In this case the algorithm does not spend too much time in bringing the nonlinear and the linearised systems close to each other. So that,unlike the case shown for θ_p=20 in Figures 5.3a and 5.3b, the objective, J, does not have a flat plateau and the gain, α, does not oscillate excessively.

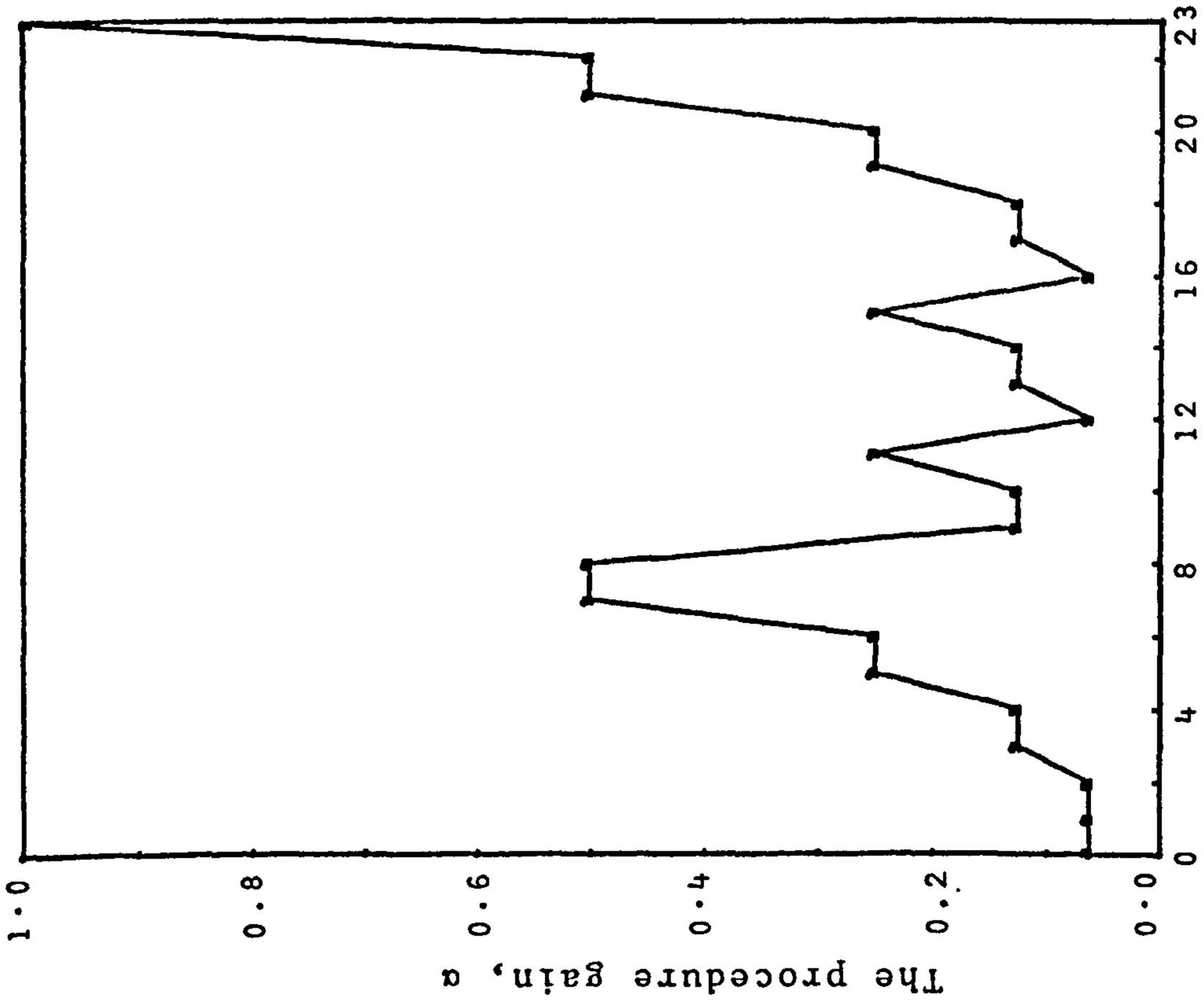


Fig.5.10b The procedure gain, α, at each iteration.
(all parameters as in Fig.5.9a)

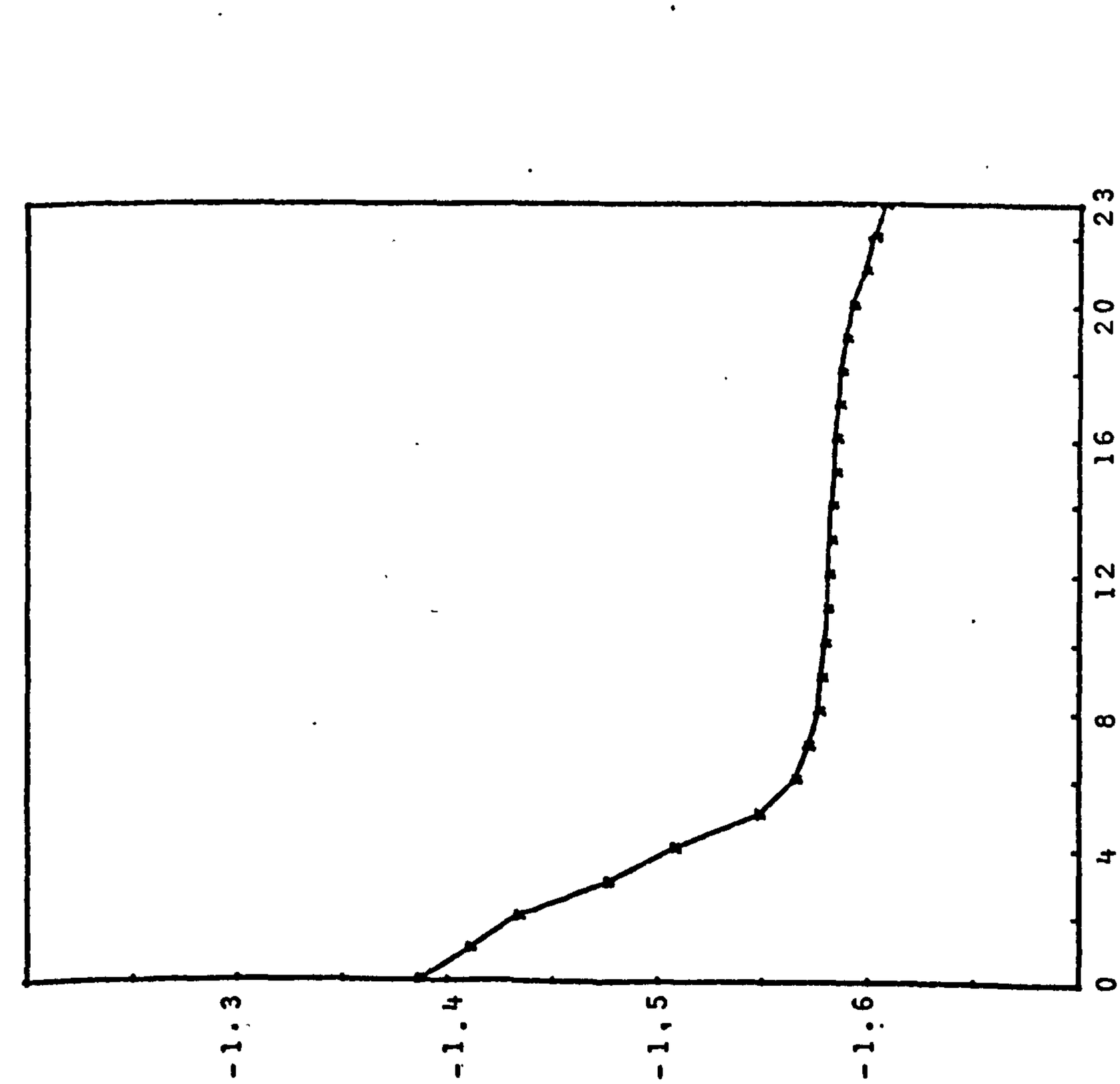


Fig.5.10a The objective function at each iteration.
(all parameters as in Fig.5.9a)

The problems examined in Cases (b) and (c) above were solved empirically in chapter 2. It was assumed before hand that the periodic inputs were on-off and unimodal in nature. The periodic performance for a given square wave was then determined by integrating the system equations forward until periodicity was established. A search on the on to off ratio and the period of the square wave was then used to find the best square wave inputs empirically. Typical results are shown in Figures 2.4 to 2.7.

In the present case, no a priory assumption about the form of the inputs is necessary. The results obtained indicated that the optimal periodic inputs were indeed of an on-off nature. In each case, the profiles obtained after a search for the optimal period, θ_p^*, were unimodal in nature and very similar to those found in chapter 2. The computation times for these runs ranged between 4 to 8 minutes depending on the period used.

The program was then used to test the optimality of the best policies obtained empirically, by feeding these policies as an initial guess for the algorithm. In all cases examined, the proposed procedure returned the same initial policy as the optimal periodic input. This fully justifies the empirical approach used in chapter 2. It also demonstrates that the proposed procedure could be used as an effective test for the optimality of a given periodic input. Naturally in practice such a test is of great importance.

Conclusions

The results obtained in this thesis indicated that the optimal enforced unsteady operation of a consecutive-competing reaction scheme in a continuous stirred tank reactor is achieved with on-off periodic inputs. Although a specific system was studied, these results apply equally well to the continuous unsteady operation of a wide range of processes with inlet control: the on-off periodic nature of the optimal inputs can be determined qualitatively by formulating the problem in accordance with optimal control theory. Such input strategies are perhaps not too difficult or expensive to implement in practice and, subject to a proper economic evaluation, may lead to significant savings.

The comparison of results from dynamic and steady modes of operation requires careful thought: it should be made under strictly comparable conditions- a point sometimes overlooked. Another fundamental point not previously considered is that the dynamic efficiency of a continuous process should be expressed as a ratio of two integral quantities: chapter 4 dealt with this point. Once the constraints which allow a strict comparison have been laid down the problem is easily formulated. However, the actual solution of the formulated problem poses considerable difficulties.

The most common approach is to assume a parametric periodic waveform for the inputs. A search technique, such as that used in chapter 2, can then easily determine the best parameters for the particular waveform chosen.

Clearly, any envisaged periodic operation must correspond to a continuous one and therefore should not contain any contributions from start-up or shut-down transients. In the above empirical approach this continuity problem is handled by measuring the performance only after a truly periodic cycle is established. Although physical reasoning may suggest the waveform to be used, there is no real guarantee that the one chosen is the best possible waveform.

The optimal control problem was solved rigorously in chapter 3 with no assumption about either the periodicity or the form of the inputs. The continuity problem mentioned above was dealt with by considering a sufficiently long operating interval for the end effects to become negligible. The final policies obtained in all cases were either steady or on-off unimodal periodic. Although the periodicity was to be expected as the process itself has no memory beyond its settling time, the unimodality was not and is perhaps a feature of the particular type of problem examined.

The continuity problem was tackled directly in the algorithm developed in chapter 5 by imposing a periodicity (but not a unimodality) condition on the inputs. This dramatically reduces the operating interval that need be considered which results in considerable savings in computational requirements. The algorithm developed makes use of a particular linearisation so that the solution of the nonlinear problem is obtained by the successive solution of a linear optimal control problem. The results

obtained using this algorithm are in agreement with those found by other methods in the thesis. A further significant application of the algorithm is as a test to determine whether a given periodic operation can be improved upon. This is easily achieved by using the given periodic inputs as the initial guess for the operating policy.

The study of continuous unsteady operation of chemical reactors is still in its infancy and opens up vast areas for application and analysis. The work presented here touches on a narrow aspect of the problem and the question of where future effort should be directed remains largely open. Much remains to be done at theoretical, experimental, and economic levels before serious consideration will be given to commercial applications of reactor operating policies of the type arrived at in this study. The inclusion of thermal effects brings about several experimental and theoretical difficulties and should be considered thoroughly. The problem of multiple periodicity has just been uncovered by Bailey [91] and throws doubt on the usual assumption of uniqueness of a periodic operation. In this respect, a great deal of work is required on the stability analysis of enforced periodic operation of nonisothermal reactors. On the optimal control side, the algorithm presented should be improved to take into account the optimality of the period and should be applied to a real reactor problem. Distributed parameter systems must also be studied from a variational point of view.

If the final goal of a commercially operated unsteady reactor appears distant, the potential rewards are on a scale that fully justifies the ever growing interest in this field.

NOTATION

A_i	concentration of reaction component, S_i,
A_{if}	input concentration of reactant, S_i,
A_{ifs}	maximum allowable value of A_{if},
C_ℓ, C_m, C_p	constants of integration,
c_p	heat capacity,
$\underline{c}$	constant vector, $\underline{c}^T=(c_1,\ldots,c_{q+2})$,
E_i	activation energy of the i^{th} reaction,
F	dynamic operation flow rate,
F_s	reference steady operation flow rate,
$H(.,.,.,.)$	Hamiltonian function,
$-\Delta H$	heat of reaction,
I_n	n x n identity matrix,
J	objective function,
$\bar{J}$	augmented objective function,
K_c	controller gain,
k_i	rate constant of the i^{th} reaction,
M_i	a set constant,
r_i	rate of the i^{th} reaction,
t	time,
t_f	operating interval,
t_p	operating period,
T	temperature,
T_f	feed temperature,
T_c	coolant temperature,
$\underline{u}$	control vector, $\underline{u}^T=(u_1,\ldots,u_r)$

U	heat transfer coefficient,
V	volume of the reactor,
w	dimensionless flow rate,
$\underline{x}$	state vector, $\underline{x}^T=(x_1,\ldots,x_n)$,
$\hat{\underline{x}}$	relaxed steady state, $\hat{\underline{x}}^T=(\hat{x}_1,\ldots,\hat{x}_n)$
x_o, x_ℓ, x_m, x_p	additional state variables,
$\underline{x}^+$	additional state vector, $\underline{x}^{+T}=(x_1^+,\ldots,x_{q+2}^+)$,
$\underline{y}$	adjoint vector, $\underline{y}^T=(y_1,\ldots,y_n)$,
y_o, y_ℓ, y_m, y_p	additional adjoint variables,
$\underline{z}$	linearised state vector, $\underline{z}^T=(z_1,\ldots,z_n)$,
$\underline{z}^+$	additional linearised state vector, $z^{+T}=(z_1^+,\ldots,z_{q+2}^+)$
α	gain of the algorithm,
α_i	dimensionless constant
$\beta(t)$	(q+2) x n matrix,
$\bar{\sigma}_i$	average overall selectivity of the i^{th} product,
θ	dimensionless time,
θ_p	dimensionless period,
$\bar{\eta}_i$	average overall yield of the i^{th} product,
ε	small positive scalar,
$\phi_1(t)$	n x n transition matrix,
$\phi_2(t)$	(q+2) x n transition matrix,
μ	on to off ratio of a binary input,
ρ	density,
τ	mean residence time,
ω	positive weighting scalar

Subscripts

"$_f$" denotes inlet conditions

"$_s$" denotes reference steady conditions

Superscripts

"$^+$" denotes additional variable

"*" denotes optimal conditions

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Appendix 1: Vector and matrix notation

Vector and matrix notation have been used extensively in this thesis. The convention adopted is as follows:

(a) All vectors are taken as column vectors and are denoted by underlined lower case letters. For instance

$$\underline{x} = \begin{bmatrix} x_1 \\ x_2 \\ \cdot \\ \cdot \\ \cdot \\ \cdot \\ x_n \end{bmatrix}$$

Components of a vector are denoted by subscripts, for example, $x_1, x_2, \ldots, x_n$, where x_i are real variables.

(b) All matrices are denoted by upper case letters, Greek letters ϕ and β, or are enclosed in pranthesis. For example

$$A = \begin{bmatrix} a_{11} & a_{12} & \cdots & a_{1n} \\ a_{21} & a_{22} & \cdots & a_{2n} \\ \cdots & \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots & \cdots \\ a_{k1} & a_{k2} & \cdots & a_{kn} \end{bmatrix}$$

where a_{ij} are real variables.

(c) The transpose of a vector or a matrix is denoted by the superscript T. For example

$$\underline{x}^T = (x_1, x_2, \ldots, x_n) \text{ and } A^T = \begin{bmatrix} a_{11} & a_{21} & \cdots & a_{k1} \\ a_{12} & a_{22} & \cdots & a_{k2} \\ \cdots & \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots & \cdots \\ a_{1n} & a_{2n} & \cdots & a_{kn} \end{bmatrix}$$

(d) Note that a column vector may be regarded as a n x 1 matrix, and a row vector is always written as $\underline{x}^T$. The transpose rule

$$(A.B)^T = B^T.A^T$$

is directly verified.

(e) The inverse of a square matrix A is denoted by A^{-1}. the following rules are easily verified

$$A^{-1}A = A\,A^{-1} = I,\quad (A^{-1})^T = (A^T)^{-1},\quad (A\ B)^{-1} = B^{-1}A^{-1}.$$

(f) When a mathematical symbol is applied to a vector or matrix, it is applied to every element of the vector or matrix. Thus we have

$$\frac{d}{dt}\underline{x} = \begin{bmatrix} \frac{d}{dt}x_1 \\ \vdots \\ \frac{d}{dt}x_n \end{bmatrix} \quad \text{and} \quad \int A\ dt = \begin{bmatrix} \int a_{11}dt & \cdots & \int a_{1n}dt \\ \vdots & \cdots & \vdots \\ \int a_{k1}dt & \cdots & \int a_{kn}dt \end{bmatrix}$$

(g) The symbol $\frac{\partial}{\partial\ \underline{x}}$ when applied to a scalar variable, say H, means the row vector

$$\frac{\partial}{\partial\ \underline{x}}H = (\frac{\partial}{\partial x_1}H, \frac{\partial}{\partial x_2}H, \ldots, \frac{\partial}{\partial x_n}H)$$

known as the gradient of function H.

(h) The symbol $\frac{\partial}{\partial\ \underline{x}}$ when applied to a vector, say $\underline{f}$, means the matrix

$$\left(\frac{\partial\ \underline{f}}{\partial\ \underline{x}}\right) = \begin{bmatrix} \frac{\partial f_1}{\partial x_1} & \cdots & \frac{\partial f_1}{\partial x_n} \\ \vdots & \cdots & \vdots \\ \frac{\partial f_k}{\partial x_1} & \cdots & \frac{\partial f_k}{\partial x_n} \end{bmatrix}$$

Appendix 2 : optimal steady operation with unrestricted inputs

The dynamics of many physical processes can be represented by a system of differential equations:

$$\frac{d\underline{x}}{dt} = \underline{f}(\underline{x},\underline{u},t) \qquad A2.1$$

where $\underline{x}^T = (x_1,\ldots,x_n)$ represents the n output or state variables, $\underline{u}^T = (u_1,\ldots,u_r)$ denotes the r input or control variables, t represents the independent variable and $\underline{f}^T(.,.,.) = (f_1(.,.,.),\ldots,f_n(.,.,.))$ is a given vector function of $\underline{x},\underline{u}$ and t.

Under steady conditions the process is described by a set of time invarient state and control variables, $\underline{x},\underline{u}$, which satisfy the algebraic equations

$$o = \underline{f}(\underline{x},\underline{u}) \qquad A2.2$$

The object is then to determine the constant controls $\underline{u}$ such that a given time invarient measure of the profit

$$J = J(\underline{x},\underline{u}), \qquad A2.3$$

is minimised. The necessary and sufficient conditions for the optimality of a steady operation were derived by Horn and Lin [47] and are presented in a generalised form below.

If the controls are unrestricted, optimal steady operation implies that

$$\frac{\partial J}{\partial \underline{u}} + \frac{\partial J}{\partial \underline{x}} \frac{\partial \underline{x}}{\partial \underline{u}} = o, \text{ r equations} \qquad A2.4$$

Now, differentiation of Eqs. (A2.2) with respect to the control vector yields

$$\frac{\partial \underline{f}}{\partial \underline{u}} + \frac{\partial \underline{f}}{\partial \underline{x}} \frac{\partial \underline{x}}{\partial \underline{u}} = o, \qquad A2.5$$

and if the matrix $(\frac{\partial \underline{f}}{\partial \underline{x}})$ is non singular $\frac{\partial \underline{x}}{\partial \underline{u}}$ can be written as

$$\frac{\partial \underline{x}}{\partial \underline{u}} = - (\frac{\partial \underline{f}}{\partial \underline{x}})^{-1} \frac{\partial \underline{f}}{\partial \underline{u}} \qquad A2.6$$

Substitution of $\frac{\partial \underline{x}}{\partial \underline{u}}$ into Eq. (A2.4) yields the necessary conditions which a steady operation must satisfy:

$$\frac{\partial J}{\partial \underline{u}} - \frac{\partial J}{\partial \underline{x}} (\frac{\partial \underline{f}}{\partial \underline{x}})^{-1} \frac{\partial \underline{f}}{\partial \underline{u}} = o, \text{ r equations} \qquad A2.7$$

As an example consider the steady operation of an isothermal stirred tank reactor with following reactions

$$S_1 \xrightarrow{r_1} S_2, \quad r_1 = k_1 \exp(-\frac{E_1}{RT}) A_1^v, \text{ a } v^{th} \text{ order reaction,}$$

$$S_1 \xrightarrow{r_2} S_3, \quad r_2 = k_2 \exp(-\frac{E_2}{RT}) A_1, \text{ a 1st order reaction}$$

The object being to maximise the yield of the desired product, S_2.

This operation is described by the equation

$$o = 1 - x_1 - \alpha_1 u x_1^v - \alpha_2 u^e x_1, \qquad A2.8$$

and the objective is given by

$$J_s = x_2 = \alpha_1 u x_1^v, \qquad A2.9$$

where

$$x_1 = A_1/A_{1f},\ x_2 = A_2/A_{1f},\ u = \exp(-E_1/RT),\ e = E_2/E_1$$

$$\alpha_1 = k\frac{V}{1F}A_{1f}^{v-1},\ \alpha_2 = k\frac{V}{2F}.$$

In this case Eq. (A3.7) becomes

$$\alpha_1 x_1^v = (\alpha_1 v u x_1^{v-1})(-1-\alpha_1 v u x_1^{v-1} - \alpha_2 u^e)^{-1}(-\alpha_1 x_1^v - \alpha_2 e u^{e-1} x_1),$$

which on rearrangement can be written as

$$\alpha_1 x_1^v\ [1-\alpha_2(ev-1)u^e] = o. \qquad 2.10$$

Then, as $x_1 = o$ is not a solution of Eq. (A2.10) it is necessary that

$$u^e = \frac{1}{\alpha_2(ev-1)} \qquad A2.11$$

Now, as $u=\exp(-E_1/RT)$ is by definition positive, Eq.(A2.11) is only meaningful if

$$ev > 1. \qquad \text{A2.12}$$

Thus, provided ev > 1, there is an optimal steady temperature which is given by

$$u^* = (\alpha_2(ev-1))^{-1/e} \qquad \text{A2.13}$$

Appendix 3 : The singular control law: derivation of Eq. (3.36)

For the problem under consideration, the state equations, the adjoint equations, and the Hamiltonian are given by Eqs. (3.27), (3.30) and (3.31) respectively. Then, for a singular control

$$\frac{\partial H}{\partial u_2} = w y_2 = o, \; y_2 = o, \qquad A3.1$$

and consequently all the time derivatives of $\frac{\partial H}{\partial u_2}$ are also zero:

$$\frac{d}{dt}\frac{\partial H}{\partial u_2} = w\dot{y}_2 = o, \; \dot{y}_2 = o. \qquad A3.2$$

Setting y_2 and $\dot{y}_2$ to zero in the adjoint system (3.30) yields

$$\frac{1}{w}\frac{d}{dt}\frac{\partial H}{\partial u_2} = \dot{y}_2 = \alpha_1 x_1 a + \alpha_2 x_3 b = o, \qquad A3.3$$

where

$$a = y_1 - y_3 \text{ and } b = y_3 - y_4. \qquad A3.4$$

It can also be easily shown that

$$\dot{a} = w + (w + \alpha_1 x_2)a - \alpha_2 x_2 b$$

$$\dot{b} = -2w + (w + \alpha_2 x_2)b \qquad A3.5$$

Differentiating Eq. (A3.3) with respect to time and using Eqs. (3.27) and (A3.5) yields

$$\frac{1}{w}\frac{d^2}{dt^2}\frac{\partial H}{\partial u_2} = \alpha_1\dot{x}_1 a + \alpha_1 x_1 \dot{a} + \alpha_2 \dot{x}_3 b + {}_2 x_3 \dot{b}$$

$$\frac{1}{w^2}\frac{d^2}{dt^2}\frac{\partial H}{\partial u_2} = \alpha_1 x_1 - 2\alpha_2 x_3 + \alpha_1 a = o \qquad \text{A3.6}$$

Equation (A3.6) does not determine u_2, so we differentiate twice more to give

$$\frac{1}{w^2}\frac{d^4}{dt^4}\frac{\partial H}{\partial u_2} = \alpha_1 \ddot{x}_1 - 2\,\alpha_2 \ddot{x}_3 + \alpha_1 \ddot{a}. \qquad \text{A3.7}$$

Now,

$$\ddot{x}_1 = -(w + \alpha_1 x_2)\dot{x}_1 - \alpha_1 x_1 \dot{x}_2,$$

$$\ddot{x}_3 = \alpha_1 x_2 \dot{x}_1 - (w+\alpha_2 x_2)\dot{x}_3 + (\alpha_1 x_1 - \alpha_2 x_3)\dot{x}_2,$$

$$\ddot{a} = (w + \alpha_1 x_2)\dot{a} - \alpha_2 x_2 \dot{b} + (\alpha_1 a - \alpha_2 b)\dot{x}_2,$$

which on substitution into Eq. (A3.7) yields, after some simplification, Eq. (3.36) which determines the singular control u_2.

Appendix 4 : <u>The general solution of linear differential equations arising in optimal control applications</u>.

Consider a linear process described by a set of differential equations

$$\frac{d}{dt}\underline{z} = F(t)\underline{z} + \underline{g}(\underline{u},t), \underline{x}(t_o) \text{ given,} \qquad A4.1$$

where $\underline{z}^T = (z_1,\ldots,z_n), \underline{u}^T = (u_1,\ldots,u_r)$, $F(t)$ is a given $n \times n$ matrix and $\underline{g}(\underline{u},t)$ is a given n-vector function.

The general solution to system (A4.1), which can be verified by direct differentiation, is given by

$$\underline{z}(t) = \phi_z(t,t_o)\left[\underline{z}(t_o) + \int_{t_o}^{t} \phi_z^{-1}(t,t_o)\underline{g}(\underline{u},t)dt\right] \qquad A4.2$$

where $\phi_z(t,t_o)$ is a $n \times n$ transition matrix defined by

$$\frac{d}{dt}\phi_z(t,t_o) = F(t)\phi_z(t,t_o), \phi_z(t_o,t_o) = I_n \qquad A4.3$$

Several easily derived properties of the transition matrix are listed below [67] :

I. If $F(t)$ is continuous, then $\phi_z(t,t_o)$ is nonsingular for all t.

II. If $F(t)$ is a constant matrix, then

$$\phi_z(t,t_o) = \exp F(t - t_o)$$

III. $\phi_z(t_1,t_o)\phi_z(t_o,t_2) = \phi_z(t_1,t_2)$,

$$\phi_z(t,t_o) = \phi_z^{-1}(t_o,t),\ \phi_z(t,t) = I_n.$$

In optimal control applications it is often necessary to consider system (A4.1) simultaneuosly with the adjoint system of differential equations:

$$\frac{d}{dt}\underline{y} = -F^T(t)\underline{y},\ \underline{y}(t_f) \text{ given,} \qquad \text{A4.4}$$

where $\underline{y}^T=(y_1,\ldots,y_n)$. The general solution to this system is given by

$$\underline{y}(t) = \phi_y(t,t_o)\underline{y}(t_o), \qquad \text{A4.5}$$

where the transition Matrix $\phi_y(t,t_o)$ is defined through

$$\frac{d}{dt}\ \phi_y(t,t_o) = -F^T(t)\phi_y(t,t_o),\phi_y(t_o,t_o) = I_n. \qquad \text{A4.6}$$

The two transition matrices $\phi_z(t,t_o)$ and $\phi_y(t,t_o)$ are related to each other through the identity

$$\phi_y^T(t,t_o)\phi_z(t,t_o) = I_n, \text{ for any } t. \qquad \text{A4.7}$$

The validity of Eq. (A4.7) is easily verified by direct differentiation using Eqs. (A4.3) and (A4.6).

However, the boundary condition on $\underline{y}(t)$ are usually specified at time t_f and Eq. (A4.5) must be modified as follows:

$$\underline{y}(t_f) = \phi_y(t_f, t_o)\underline{y}(t_o),$$

$$\underline{y}(t_o) = \phi_y^{-1}(t_f, t_o)\underline{y}(t_f),$$

so that Eq. (A4.5) becomes

$$\underline{y}(t) = \phi_y(t, t_o)\ \phi_y^{-1}(t_f\ , t_o)\underline{y}(t_f) \qquad \text{A4.8}$$

Then, using the properties of transition matrices, Eq. (A4.8) can be simplified to

$$\underline{y}(t) = \phi_y(t, t_o)\phi_y(t_o, t_f)\underline{y}(t_f) \cdot$$

$$= \phi_y(t, t_o)\phi_y(t_o, t)\phi_y(t, t_f)\underline{y}(t_f)$$

$$= \phi_y^{-1}(t_f, t)\underline{y}(t_f) \qquad \text{A4.9}$$

It remains to determine $\phi_y^{-1}(t_f, t)$. Consider the identity

$\phi_y(t_f, t)\phi_y(t, t_f) = I_n$,
differentiating both sides with respect to time yields:

$$\phi_y(t_f, t)\phi_y(t, t_f) + \phi_y(t_f, t)\phi_y(t, t_f) = o. \qquad \text{A4.10}$$

Then, recalling that

$$\dot{\phi}_y(t, t_f) = \dot{\phi}_y(t, t_o)\phi_y(t_o, t_f),$$

and using Eq. (A4.6) and the properties of transition matrices we arrive at

$$\frac{d}{dt}\,\phi_y(t_f,t) = \phi_y(t_f,t)F^T(t), \phi_y(t_f,t_f) = I_n. \qquad A4.11$$

Next, consider the identity

$$\phi_y(t_f,t)\phi_y^{-1}(t_f,t) = I_n,$$

and differentiate either side with respect to time to give

$$\dot{\phi}_y(t_f,t)\phi_y^{-1}(t_f,t) + \phi_y(t_f,t)\dot{\phi}_y^{-1}(t_f,t) = o.$$

Then, using Eq. (A4.11) we arrive at

$$\frac{d}{dt}\,\phi_y^{-1}(t_f,t) = -F^T(t)\phi_y^{-1}(t_f,t), \phi_y^{-1}(t_f,t_f) = I_n, \qquad A4.12$$

which together with Eq. (A4.9) completes the general solution of system (A4.4) with the boundary conditions given at time t_f. Now, noting that $(A^T)^{-1} = (A^{-1})^T$, from Eq. (A4.7) we have

$$\begin{aligned}\phi_z(t,t_o) &= \phi_y^{-1}(t,t_o)^T \\ &= \phi_y^{-1}(t_f,t_o)\phi_y^{-1}(t,t_f)^T \\ &= \phi_y^T(t_f,t).\ \phi_y^T(t_f,t_o)^{-1}\end{aligned} \qquad A4.13$$

and

$$\phi_z^{-1}(t,t_o) = \phi_y^T(t_f,t_o).\ \phi_y^{-1}(t_f,t)^T. \qquad A4.14$$

Substituting for $\phi_z(t,t_o)$ and $\phi_z^{-1}(t,t_o)$ into Eqs. (A4.2) yields

$$\underline{z}(t) = \phi_y^T(t_f,t)\left[\phi_y^{-1}(t_f,t_o)^T\ \underline{z}(t_o) + \int_{t_o}^{t} \phi_y^{-1}(t_f,t)^T\ \underline{g}(\underline{u},t)dt.\right] \qquad \text{A4.15}$$

Finally, introducing a transition matrix $\phi(t) = \phi_y^{-1}(t_f,t)^T$:

$$\frac{d\phi(t)}{dt} = -\ \phi(t)\ F(t),\ \phi(t_f) = I_n, \qquad \text{A4.16}$$

from equations (A4.9), (A4.12) and (A4.15) the general solution to systems

$$\frac{d}{dt}\ \underline{z}(t) = F(t)\underline{z}(t) + \underline{g}(\underline{u}(t),t),\ \underline{z}(t_o)\ \text{given}, \qquad \text{A4.17}$$

and

$$\frac{d}{dt}\ \underline{y}(t) = F^T(t)\underline{y}(t),\ \underline{y}(t_f)\ \text{given}, \qquad \text{A4.18}$$

can be written down as

$$\underline{z}(t) = \phi^{-1}(t)\left[\phi(t_o)\underline{z}(t_o) + \int_{t_o}^{t} \phi(t)\underline{g}(\underline{u}(t),t)dt.\right] \qquad \text{A4.19}$$

$$\underline{y}(t) = \phi^T(t)\underline{y}(t_f). \qquad \text{A4.20}$$

Appendix 5: A program for determination of optimal periodic input profiles

The program listed below is a realisation of the algorithm developed in chapter 5. A standard variable step fourth order Runge-Kutta method is used for all numerical integrations, the numerical quadratures are carried out by the repeated application of a five point closed Newton-Coates formula, and the matrix inversions are performed by Gaussian elimination using the maximum pivot strategy. These numerical methods are fully described in [92].

A listing of the principal variables used in the program is given below:

MODE	≥ 0, for integral objectives < 0, for ratio-integral objectives
N	number of state variables, n
IR	number of control variables, r
IQ	number of constrains, q. The program sets it to q+1 if MODE ≥ 0 q+2 if MODE < 0
TP	period, t_p
TDEL	discretisation interval, Δt. The program adjusts this to suit the quadrature formula used.
NP	number of storage points
ERR	error bound on the integrations
WEIGHT	positive weighting scalar, ω
UT(IR,NP)	$\underline{u}(t)$, $0 \leq t \leq t_p$
XT(N,NP)	$\underline{x}(t)$, $0 \leq t \leq t_p$

B1T(N,N,NP)	$\phi_1(t)$, $0 \leqslant t \leqslant t_p$
B2T(IQ,N,NP)	$\phi_2(t)$ or $\beta(t)$, $0 \leqslant t \leqslant t_p$
US(IR,NP)	$\underline{v}^*(t)$, $0 \leqslant t \leqslant t_p$
U(IR)	$\underline{u}(t)$, at a given time t
X(N)	$\underline{x}(t)$, at a given time t
B(IQ,N)	$\beta(t)$, at a given time t
UMAX(IR)	$\underline{v}^*(t)$, at a given time t
F(N)	$\underline{f}(\underline{x},\underline{u})$, at a given time t
FP(IQ)	$\underline{f}^+(\underline{x},\underline{u})$, at a given time t
DFDX(N,N)	$\partial \underline{f}/\partial \underline{x}$ at a given time t
DFPDX(IQ,N)	$\partial \underline{f}^+/\partial \underline{x}$ at a given time t
G(N)	$\underline{f}(\underline{x},\underline{v})-(\partial \underline{f}/\partial \underline{x})\underline{x}$ at a given time t
GP(IQ)	$\underline{f}^+(\underline{x},\underline{v})-(\partial \underline{f}^+/\partial \underline{x})\underline{x}$ at a given time t
ALFA	the gain, α
E	the scalar search variable, ε
XP(IQ)	$\underline{x}^+(t_p)$
XPL(IQ)	$\underline{z}^+(t_p)$
XO(N)	the periodic end state, $\underline{x}(0)=\underline{x}(t_p)$
XS(N)	the periodic end state, $\underline{z}^*(0)=\underline{z}^*(t_p)$

The program is user orientated and requires the following data:

1: t_p, Δt, err
2: MODE, q
3: $M_1, M_2, .., M_q$
4: ω
5: n,r

The subroutine GUESS is used to obtain an initial control policy, $\underline{u}(t)$, $0 \leqslant t \leqslant t_p$, and an initail starting point for the process, $\underline{x}(0)$. The user should also supply subroutines which,

given $\underline{\dot{x}}(t)$, $\underline{u}(t)$, and $\beta(t)$ at a given time t, return the values of $\underline{f}(\underline{x}(t),\underline{u}(t))$, $\underline{f}^{+}(\underline{x}(t),\underline{u}(t))$, $(\partial\underline{f}/\partial\underline{x})$, $(\partial\underline{f}^{+}/\partial\underline{x})$, and the control vector $\underline{v}^{*}(t)$ which maximises the Hamiltonian for the linearised system: $h= \underline{c}^{T}[\beta(t)\underline{f}(\underline{x},\underline{v})+\underline{f}^{+}(\underline{x},\underline{v})]$. The format of these subroutines is given in the program listing below.

The program is written for control constraints of the type

$$0 \leqslant u_i(t) \leqslant 1, \text{ for all } t, \ i=1,..,r.$$

Constraints of the type

$$u_i^{min} \leqslant u_i(t) \leqslant u_i^{max}, \text{ for all } t, \ i=1,...,r,$$

should first be converted to above form by means of the substitution: $u_i^{\circ}(t)=(u_i(t)-u_i^{min})/(u_i^{max}-u_i^{min})$, $i=1,...,r$. To use the program with other types of control constraints the subroutine returning $\underline{v}^{*}(t)$ should be written according to the type of control constraint in force.

```
C
C     MAIN PROGRAM:   A GENERAL ALGORITHM FOR DETERMINATION OF
C                     OPTIMAL PERIODIC INPUT PROFILES.
C
      IMPLICIT REAL*8 (A-H,O-Z)
      REAL*8 JC,JBO,JL,JBL,JH,JBH,J1,JB1
      DIMENSION X0(4),X(4),XS(4),V(4),F(4),FL(4),U(1),UMAX(1),C(3),XP(3)
     1,XPO(3),XPL(3),XPH(3),FP(3),FPL(3),P10(4,4),DFDX(4,4),V1(4,4),P20(
     23,4),DFDX(3,4),B(3,4),V2(3,4),SUM1(3,5),SUM2(4,5),PRMT(5),AUX(8,4
     3),AUXP(8,28),P(28),DP(28),U0(1,81),US(1,81),G(4),GP(3),XH(4),YP(3)
      DIMENSION YP1(3),XPL1(3)
      COMMON /Z1/ UT(1,81)
      COMMON /Z2/ XT(4,81)
      COMMON /Z3/ B1T(4,4,81)
      COMMON /Z4/ B2T(3,4,81)
C
      EXTERNAL FCTX,OUTX,FPNLN,FCTP,OUTP,FCTXL
C
C     READ FORMATS
C
  100 FORMAT(5D15.8)
  101 FORMAT(5I5)
C
C     WRITE FORMATS
C
  200 FORMAT(1H ,80(1H*)//1H ,'PERIOD=',D13.6,2X,'STORAGE INT=',D13.6,
     12X/1H ,'ERROR CRITERIA=',D13.6,3X,I4,2X,'STORAGE POINTS'//)
  201 FORMAT(1H ,'ORDINARY INTEGRAL OBJECTIVE'/)
  202 FORMAT(1H ,'RATIO-INTEGRAL OBJECTIVE'/)
  203 FORMAT(1H ,'NO INTEGRAL SIDE CONSTRAINTS'/)
  204 FORMAT(1H ,I4,2X,'INTEGRAL SIDE CONSTRAINTS:'/)
  205 FORMAT(1H ,5(D13.6,2X)//)
  206 FORMAT(1H ,I4,2X,'STATE VARIABLES',I4,2X,'CONTROL VARIABLES'//1H ,
     280(1H*)//////////)
  207 FORMAT(1H ,'WEIGHTING SCALAR IS  ',D13.6/)
C
C       READ AND WRITE INITIAL DATA FOR THE RUN
C
      READ(5,100) TP,TDEL,ERR
      NP=TP/TDEL+0.500
      I=NP/4
      MP=4*I
      IF(NP-MP.EQ.0) GO TO 1
      NP=MP
      TDEL=TP/NP
    1 NP=NP+1
      WRITE(6,200) TP,TDEL,ERR,NP
      READ(5,101) MODE,IQ
      IF(MODE)2,2,3
    2 WRITE(6,201)
      GO TO 4
    3 WRITE(6,202)
    4 IF(IC.GT.0) GO TO 5
      WRITE(6,203)
      GO TO 6
    5 WRITE(6,204) IQ
      READ(5,100) (C(I),I=1,IQ)
      WRITE(6,205) (C(I),I=1,IQ)
      READ(5,100) WEIGHT
      WRITE(6,207) WEIGHT
    6 IF(MODE) 7,7,8
    7 IQ=IC+1
      GO TO 9
    8 IQ=IC+2
    9 READ(5,101) N,IR
      WRITE(6,206) N,IR
      NC=N+IC
      NN=N*NP
      WTT=1.D0/NN
      WT=1.D0/N
C
C     GUESS AN INITIAL STATE X(0) AND A CONTROL PROFILE OVER
C     ONE PERIOD.
C
      CALL GUESS(X0,N,IR,NP,TP,TDEL)
      DO 10 I=1,IR
      DO 10 J=1,NP
   10 U0(I,J)=UT(I,J)
C
C
C     START THE ITERATIVE LOOP.
C
      ALFA=0.0625D0
      E=1.D0
      IT=0
      PRMT(1)=0.D0
      PRMT(2)=TP
      PRMT(3)=TDEL
      PRMT(4)=ERR
      IP=1
   11 DO 12 I=1,N
      X(I)=X0(I)
   12 F(I)=WT
C
C     INTEGRATE THE NONLINEAR SYSTEM EQUATIONS FORWARD
C     UNTIL PERIODICITY IS ESTABLISHED
C
      CALL DRKGS(PRMT,X,F,N,IHLF,FCTX,OUTX,AUX,U,IR,V,N,V1,V2,IC)
C
      S=0.D0
```

```
01010       DO 12 I=1,N
01020    13 S=S+(X(I)-XO(I))**2
01030       IF(DSQRT(S).LE.ERR) GO TO 15
01040       IP=IP+1
01050       DO 14 I=1,N
01060    14 XO(I)=X(I)
01070       GO TO 11
01080    15 DO 16 I=1,N
01090    16 XO(I)=X(I)
01100 C
01110 C     ESTIMATE THE OBJECTIVE AND THE CONSTRAINTS
01120 C     ONE PERIOD.
01130 C
01140       CALL OBJCON(PRMT,XPO,FP,IQ,X,N,U,IR,SUM1,FPNLN)
01150       IF(IT.GE.1) GO TO 77
01160       L=IC-1
01170       M=IC-2
01180       IF(MODE.GT.0) GO TO 75
01190       IF(L.GT.0) GO TO 76
01200       GO TO 77
01210    76 DO 78 I=1,L
01220    78 YP(I)=0.DO
01230       GO TO 77
01240    75 IF(M.LE.0) GO TO 79
01250       DO 80 I=1,M
01260    80 YP(I)=0.DO
01270    79 YP(L)=XPO(IQ)/XPO(L)
01280    77 CONTINUE
01290       CALL OBJECT(PRMT,XPO,IQ,C,D,MODE,JC,JBO,WEIGHT,YP)
01300 C
01310 C
01320 C     WRITE THE RESULTS OF ITERAION.
01330 C
01340       ITEST=-1
01350    17 ITEST=ITEST+1
01360       CALL PNLIN(PRMT,XC,N,XPO,IQ,C,D,JO,JBO,NP,IR,MODE,ALFA,IT)
01370 C
01380 C     DOUBLE THE GAIN (ALFA) AFTER TWO SUCCESSIVE
01390 C     SUCCESSFUL ITERATIONS
01400 C
01410       IF(ITEST.LT.2) GO TO 73
01420       ALFA=ALFA+ALFA
01430       IF(ALFA.GT.1.DO) ALFA=1.DO
01440       ITEST=0
01450    73 CONTINUE
01460 C
01470 C     START OF THE INNER ITERATIVE LOOP
01480 C
01490       PRMT(1)=TP
01500       PRMT(2)=0.DO
01510       PRMT(3)=-TDEL
01520       DO 18 I=1,NQ
01530       DO 18 J=1,N
01540       K=I+(J-1)*NQ
01550       P(K)=0.DO
01560    18 DP(K)=ATT
01570       DO 19 I=1,N
01580    19 P(I+(I-1)*NQ)=1.DO
01590 C
01600 C     CALCULATE THE TRANSITION MATRICES (PHI1 AND PHI2)
01610 C
01620       CALL DRKGS(PRMT,P,DP,NN,IHLF,FCTP,CUTP,AUXP,U,IR,V,N,DFDX,DFDPDX,IQ
01630      1)
01640 C
01650 C     STORE PHI1(0) AND PHI2(0).
01660 C
01670       DO 20 I=1,N
01680       DO 20 J=1,N
01690    20 P10(I,J)=B1T(I,J,1)
01700       DO 21 I=1,IQ
01710       DO 21 J=1,N
01720    21 P20(I,J)=B2T(I ,J,1)
01730 C
01740 C     FIND THE INVERSE OF I-PHI1(0) AND DETERMINE THE MATRIX BETA
01750 C
01760       CALL INVERT(P10,N,V1)
01770 C
01780       DO 23 I=1,IQ
01790       DO 23 J=1,N
01800       S=0.DO
01810       DO 22 K=1,N
01820    22 S=S+P20(I,K)*V1(K,J)
01830    23 V2(I,J)=S
01840       DO 25 L=1,NP
01850       DO 25 I=1,IQ
01860       DO 25 J=1,N
01870       S=0.DO
01880       DO 24 K=1,N
01890    24 S=S+V2(I,K)*B1T(K,J,L)
01900    25 B2T(I,J,L)=B2T(I,J,L)+S
01910 C
01920 C     START OF THE SEARCH FOR THE OPTIMAL CONTROLS FOR
01930 C     THE LINEARISED SYSTEM.
01940 C
01950       ITL=1
01960       ITST=0
01970       E=1.D0
01980       DO 26 I=1,IQ
01990       YP1(I)=YP(I)
02000    26 XPL1(I)=XPO(I)
```

```
C
C     ADJUST THE VALUES OF THE CONSTANT C1,C2,...
C
   27 CALL YPLUS(YP,XPL1,C,E,IC,ITL,MODE,YP1)
C
C     WITH THE ESTIMATED VALUES OF THE ADJOINT CONSTANTS
C     FIND THE OBJECTIVE AND THE INTEGRAL SIDE CONSTRAINTS
C     WITH A CONTROL VECTOR WHICH MAXIMISES THE HAMILTONIAN
C     AT EACH POINT IN TIME.
C
      DO 28 I=1,IQ
      DO 28 J=1,5
   28 SUM1(I,J)=0.D0
      K=1
   29 DO 30 I=1,IP
   30 U(I)=UC(I,K)
      DO 31 I=1,N
   31 X(I)=XT(I,K)
      T=(K-1)*TDEL
      DO 32 I=1,IQ
      DO 32 J=1,N
   32 B(I,J)=BZT(I,J,K)
      CALL MAXHAM(T,X,N,U,IR,F,FP,IQ,YP,B,UMAX)
      DO 33 I=1,IR
   33 UT(I,K)=UMAX(I)
      CALL GLIN(T,X,N,U,UMAX,IR,F,DFDX,G,IQ)
      CALL GPLIN(T,X,N,U,UMAX,IR,FP,DFPDX,GP,IQ)
      DO 35 I=1,IQ
      S=0.D0
      DO 34 J=1,N
   34 S=S+B(I,J)*G(J)
   35 FPL(I)=S+GP(I)
      DO 36 I=1,IQ
   36 SUM1(I,1)=SUM1(I,1)+FPL(I)
      IF(K.GE.NP) GO TO 37
      K=NP
      GO TO 29
   37 DO 44 M=2,5
      L=NP+M-5
      IF(M.EQ.5) L=L-4
      DO 44 K=M,L,4
      DO 38 I=1,IP
   38 U(I)=UC(I,K)
      DO 39 I=1,N
   39 X(I)=XT(I,K)
      T=(K-1)*TDEL
      DO 40 I=1,IQ
      DO 40 J=1,N
   40 B(I,J)=BZT(I,J,K)
C
      CALL MAXHAM(T,X,N,U,IR,F,FP,IQ,YP,B,UMAX)
      DO 41 I=1,IR
   41 UT(I,K)=UMAX(I)
      CALL GLIN(T,X,N,U,UMAX,IR,F,DFDX,G,IQ)
      CALL GPLIN(T,X,N,U,UMAX,IR,FP,DFPDX,GP,IQ)
C
      DO 43 I=1,IQ
      S=0.D0
      DO 42 J=1,N
   42 S=S+B(I,J)*G(J)
   43 FPL(I)=S+GP(I)
      DO 44 I=1,IQ
   44 SUM1(I,M)=SUM1(I,M)+FPL(I)
      CC=2.D0*TDEL/45.D0
      DO 45 I=1,IQ
      XPL(I)=7.D0*(SUM1(I,1)+2.D0*SUM1(I,5))+12.D0*SUM1(I,3)
   45 XPL(I)=CC*(XPL(I)+32.D0*(SUM1(I,2)+SUM1(I,4)))
C
C     FIND THE OBJECTIVE AND THE CONSTRAINTS FOR THE
C     LINEARISED SYSTEM OVER ONE PERIOD.
C
      CALL OBJECT(PRMT,XPL,IQ,C,D,MODE,JL,JBL,WEIGHT,YP)
C
C     ADJUST E UNTIL A BETTER SET OF C1,C2,... IS OBTAINED
C
      IF(MODE) 46,46,47
   46 IF(IC.LT.2) GO TO 53
   47 IF(ITL.GT.1) GO TO 50
   48 JL1=JL
      JBL1=JBL
      DO 81 I=1,IQ
      YP1(I)=YP(I)
   81 XPL1(I)=XPL(I)
C
C     WRITE THE RESULTS FOR THE LINEARISED SYSTEM.
C
   49 CALL RLIN(PRMT,XS,N,XPL,YP,C,IQ,D,JL,JBL,NP,IR,E,ITL,ITST)
      ITL=ITL+1
      GO TO 27
   50 IF(JBL.LE.JBL1) GO TO 51
C
C     HALVE E FOR EACH UNSUCCESSFUL STEP
C
      ITST=0
      E=E/2.D0
      GO TO 49
C
C     DOUBLE E AFTER 2 SUCCESSIVE SUCCESSFUL STEPS.
C
   51 IF(DABS(JBL1-JBL).LE.1.D-05.AND.D.LE.1.D-06) GO TO 53
```

```
      IF(CABS(JBL-JBL).LE.1.D-05.AND.E.LE.1.D-06) GO TO 53
      ITST=ITST+1
      IF(ITST.GT.2) GO TO 52
      GO TO 48
   52 E=E+E
      GO TO 48
C
C     RECORD THE OPTIMAL CONTROL FOR THE LINEARISED SYSTEM,
C     AND FIND THE CORRESPONDING OPTIMAL PERIODIC END STATES.
C
   53 DO 54 I=1,IR
      DO 54 J=1,NP
   54 US(I,J)=LT(I,J)
C
      CALL QBODF(PRMT,XS,FL,N,X,N,U,IR,SUM2,FCTXL)
C
      DO 56 I=1,N
      S=0.D0
      DO 55 J=1,N
   55 S=S+V1(I,J)*XS(J)
   56 V(I)=S
      DO 57 I=1,N
   57 XS(I)=V(I)
C
C     END OF INNERMOST ITERATIVE LOOP
C
      ITL=-ITL
C
      CALL ELIN(PRMT,XS,N,XFL,YP,C,IC,D,JL,JBL,NP,IR,E,ITL,ITST)
C
C     DETERMINE AN INTERMEDIATE CONTROL WHICH IMPROVES
C     THE NONLINEAR SYSTEM.
C
   58 AL=1.D0-ALFA
      DO 59 I=1,IR
      DO 59 J=1,NP
   59 LT(I,J)=AL*U0(I,J)+ALFA*US(I,J)
      DO 60 I=1,N
   60 XH(I)=AL*X0(I)+ALFA*XS(I)
C
C     INTEGRATE THE NONLINEAR SYSTEM EQUATIONS FORWARD
C     UNTIL PERIODICITY IS ESTABLISHED
C
      PRMT(1)=0.D0
      PRMT(2)=TP
      PRMT(3)=TDEL
      IP=1
   61 DO 62 I=1,N
      X(I)=XH(I)
   62 F(I)=WT
C
      CALL DRKGS(PRMT,X,F,N,IHLF,FCTX,OUTX,AUX,U,IR,V,N,V1,V2,IQ)
C
      S=0.D0
      DO 63 I=1,N
   63 S=S+(X(I)-XH(I))**2
      IF(DSQRT(S).LE.ERR) GO TO 65
      IP=IP+1
      DO 64 I=1,N
   64 XH(I)=X(I)
      GO TO 61
   65 DO 66 I=1,N
   66 XH(I)=X(I)
C
C     ESTIMATE THE OBJECTIVE AND THE CONSTRAINTS OVER
C     ONE PERIOD.
C
      CALL QBODF(PRMT,XPH,FP,IC,X,N,U,IR,SUM1,FPNLN)
C
      CALL OBJECT(PRMT,XPH,IQ,C,D,MODE,JL,JBH,WEIGHT,YP)
C
C     ADJUST ALFA UNTIL A BETTER RESULT IS OBTAINED.
C
      IF(JBH.GT.JB0) GO TO 70
      DO 67 I=1,IR
      DO 67 J=1,NP
   67 U0(I,J)=LT(I,J)
      IF(ALFA.GE.1.D0) GO TO 71
   83 DO 68 I=1,N
   68 X0(I)=XH(I)
      DO 69 I=1,IQ
   69 XP0(I)=XPH(I)
      J0=JH
      JB0=JBH
      IT=IT+1
      IF(IT.GT.101) GO TO 71
      GO TO 17
C
C     HALVE ALFA FOR UNSUCCESSFUL STEPS
C
   70 ALFA=ALFA/2.D0
      ITEST=0
      GO TO 58
C
C     TEST FOR CONVERGENCE TO THE OPTIMAL NONLINEAR CONTROL
C
   71 S=0.D0
      DO 82 I=1,N
   82 S=S+(X0(I)-XH(I))**2
      IF(DSQRT(S).LE.ERR) GO TO 72
```

```
      GO TO 83
C
C     WRITE THE FINAL RESULTS OF THE RUN
C
   72 CALL FNLIN(PRMT,XH,N,XPH,IQ,C,D,JH,JBH,NP,IR,MODE,ALFA,IT)
C
      STOP
      END
      SUBROUTINE FNLN(T,X,N,U,IR,F,IQ)
      IMPLICIT REAL*8 (A-H,O-Z)
      DIMENSION X(N),U(IR),F(N)
C
C    USER SUPPLIED: GIVEN  X VECTOR AND U VECTOR IT SHOULD
C     RETURN THE VECTOR F(X,U).
C
      W=1.00/0.55D0
      A1=4.0DC
      A2=A1
      R1=A1*X(1)*X(2)
      R2=A2*X(2)*X(3)
      F(1)=W*(U(1)-X(1))-R1
      F(2)=W*(U(1)-X(2))-R1-R2
      F(3)=-W*X(3)+R1-R2
      F(4)=-W*X(4)+R2
C
      RETURN
      END
      SUBROUTINE FPALN(T,X,N,U,IR,FP,IQ)
      IMPLICIT REAL*8 (A-H,O-Z)
      DIMENSION X(N),U(IR),FP(IQ)
C
C     USER SUPPLIED: GIVEN THE X VECTOR AND THE U VECTOR IT
C     SHOULD RETURN THE VECTOR F+(X,U)
C
      W=1.00/0.55D0
      FP(1)=W*U(1)
      FP(2)=W*(U(1)-X(1))
      FP(3)=-W*X(3)
C
      RETURN
      END
      SUBROUTINE DFDXT(T,X,N,U,IR,DFDX)
      IMPLICIT REAL*8 (A-H,O-Z)
      DIMENSION X(N),U(IR),DFDX(N,N)
C
C    USER SUPPLIED: GIVEN THE X VECTOR AND THE U VECTOR IT
C    SHOULD RETURN THE MATRIX  DF(X,U)/DX
C
      W=1.00/0.55D0
      A1=4.0DC
      A2=A1
      R1=A1*X(1)
      R2=A1*X(2)
      R3=A2*X(2)
      R4=A2*X(3)
      DO 1 I=1,N
      DO 1 J=1,N
    1 DFDX(I,J)=0.D0
      DFDX(1,1)=-W-R2
      DFDX(1,2)=-R1
      DFDX(2,1)=-R2
      DFDX(2,2)=-W-R1-R4
      DFDX(2,3)=-R3
      DFDX(3,1)=R2
      DFDX(3,2)=R1-R4
      DFDX(3,3)=-W-R3
      DFDX(4,2)=R4
      DFDX(4,3)=R3
      DFDX(4,4)=-W
C
      RETURN
      END
      SUBROUTINE DFPDXT(T,X,N,U,IR,DFPDX,IQ)
      IMPLICIT REAL*8 (A-H,O-Z)
      DIMENSION X(N),U(IR),DFPDX(IQ,N)
C
C     USER SUPPLIED: GIVEN THE X VECTOR AND THE U VECTOR IT
C     RETURN THE MATRIX DF+(X,U)/DX
C
      W=1.00/0.55D0
      DO 1 I=1,IQ
      DO 1 J=1,N
    1 DFPDX(I,J)=0.DO
      DFPDX(2,1)=-W
      DFPDX(3,3)=-W
C
      RETURN
      END
      SUBROUTINE MAXHAM(T,X,N,U,IR,F,FP,IQ,YP,B,UMAX)
      IMPLICIT REAL*8 (A-H,O-Z)
      DIMENSION X(N),U(IR),F(N),FP(IQ),YP(IQ),B(IQ,N),UMAX(IR)
C
C     USER SUPPLIED: GIVEN THE X VECTOR, THE U VECTOR, THE MATRIX
C     BETA, AND A CONSTANT VECTOR C (YP IN THE PROGRAM) AT A GIVEN
C     TIME T, IT SHOULD RETURN THE VECTOR UMAX WHICH YIELDS THE
C     HIGHEST VALUE OF THE HAMILTONIAN FOR THE LINEARISED SYSTEM
C
      DIMENSION Q(4,2),CP(3,2),IU(2),A(3,2),C(2)
      W=1.00/0.55D0
      DO 7 I=1,N
```

```
      DO 7 J=1,IR
    7 Q(I,J)=C.DO
      C(1,1)=h
      C(2,1)=h
      DO 8 I=1,IQ
      DO 8 J=1,IR
    8 CP(I,J)=C.DO
      CP(1,1)=h
      CP(2,1)=h
      DO 1 I=1,IR
    1 IU(I)=1
      DO 3 I=1,IQ
      DO 3 J=1,IR
      S=0.DO
      DO 2 K=1,N
    2 S=S+B(I,K)*Q(K,J)
    3 A(I,J)=S+CP(I,J)
      DO 6 J=1,IR
      S=0.DO
      DO 4 I=1,IQ
    4 S=S+YP(I)*A(I,J)
      IF(S)5,5,6
    5 IU(J)=-1
    6 UMAX(J)=(1+IU(J))/2
C
      RETURN
      END
      SUBROUTINE GUESS(XO,N,IR,NP,TP,TDEL)
      IMPLICIT REAL*8 (A-H,C-Z)
      DIMENSION XO(N)
      COMMON /Z1/ UT(1,81)
C
C     USER SUPPLIED: SHOULD RETURN AN INITIAL CONTROL POLICY
C     AND AN INITIAL STATE
C
      DO 1 I=1,N
    1 XO(I)=5.C-01
      DO 2 I=1,IR
      DO 2 J=1,NP
      XX=(J-1)*TDEL*3.14159265DO/TP
    2 UT(I,J)=C.5DO*(1.DO+DSIN(XX))
C
      RETURN
      END
      SUBROUTINE OBCDE(PRMT,XA,FA,NA,X,N,U,IR,SUM,FUNC)
      IMPLICIT REAL*8 (A-H,C-Z)
      DIMENSION XA(NA),FA(NA),X(N),U(IR),SUM(NA,5),PRMT(5)
      COMMON /Z1/ UT(1,81)
      COMMON /Z2/ XT(4,81)
      COMMON /Z3/ BIT(4,4,81)
      COMMON /Z5/ IT
      ITM=(PRMT(2)-PRMT(1))/PRMT(3)+1.00002DO
      DO 1 I=1,NA
      DO 1 J=1,5
    1 SUM(I,J)=0.DO
      IT=1
    2 DO 3 I=1,IR
    3 U(I)=UT(I,IT)
      DO 4 I=1,N
    4 X(I)=XT(I,IT)
      T=(IT-1)*DABS(PRMT(3))
      CALL FUNC(T,X,N,U,IR,FA,NA)
      DO 5 I=1,NA
    5 SUM(I,1)=SUM(I,1)+FA(I)
      IF(IT.GE.ITM) GO TO 6
      IT=ITM
      GO TO 2
    6 DO 9 J=2,5
      L=ITM+J-5
      IF(J.EQ.5) L=L-4
      DO 9 IT=J,L,4
      DO 7 I=1,IR
    7 U(I)=UT(I,IT)
      DO 8 I=1,N
    8 X(I)=XT(I,IT)
      T=(IT-1)*DABS(PRMT(3))
      CALL FUNC(T,X,N,U,IR,FA,NA)
      DO 9 I=1,NA
    9 SUM(I,J)=SUM(I,J)+FA(I)
      C=2.DO*DABS(PRMT(3))/45.DO
      DO 10 I=1,NA
      XA(I)=7.DO*(SUM(I,1)+2.DO*SUM(I,5))+12.DO*SUM(I,3)
   10 XA(I)=(XA(I)+32.DO*(SUM(I,2)+SUM(I,4)))*C
   11 RETURN
      END
      SUBROUTINE OBJECT(PRMT,XP,IQ,C,D,MODE,XJ,XBJ,WEIGHT,YP)
      IMPLICIT REAL*8 (A-H,C-Z)
      DIMENSION PRMT(5),XP(IQ),C(IQ),YP(IQ)
C
      IF(MODE) 3,3,1
    1 C(IQ)=XP(IQ)
      C(IQ-1)=XP(IQ-1)
      IF(XP(IQ-1).EQ.0.DO) GO TO 2
      XJ=XP(IQ)/XP(IQ-1)
      GO TO 4
    2 XJ=1.D+6C
      WRITE(6,100)
  100 FORMAT(1H ,'WARNING ZERO DENOMINATOR')
      GO TO 4
    3 C(IQ)=XP(IQ)
```

```
      XJ=XP(IC)/DABS(PRMT(2)-PRMT(1))
    4 D=0.D0
      DO 5 I=1,IQ
    5 D=D+(XP(I)-C(I))**2
      IF(NCDF.GT.0) GO TO 7
      XBJ=WEIGHT*D/2.D0+XJ
      RETURN
    7 XBJ=(YP(IQ-1)-XJ)**2+WEIGHT*D/2.D0
C
      RETURN
      END
      SUBROUTINE GLIN(T,X,N,L,UMAX,IR,F,DFDX,G,IQ)
      IMPLICIT REAL*8 (A-H,O-Z)
      DIMENSION X(N),U(IR),UMAX(IR),F(N),DFDX(N,N),G(N)
C
      CALL FNLN(T,X,N,UMAX,IR,F,IQ)
C
      CALL DFDXT(T,X,N,U,IR,DFDX)
C
      DO 2 I=1,N
      S=0.D0
      DO 1 J=1,N
    1 S=S+DFDX(I,J)*X(J)
    2 G(I)=F(I)-S
      RETURN
      END
      SUBROUTINE GPLIN(T,X,N,U,UMAX,IR,FP,DFPDX,GP,IQ)
      IMPLICIT REAL*8 (A-H,O-Z)
      DIMENSION X(N),U(IR),UMAX(IR),FP(IQ),DFPDX(IQ,N),GP(IQ)
C
      CALL FPLN(T,X,N,UMAX,IR,FP,IQ)
C
      CALL DFPDXT(T,X,N,U,IR,DFPDX,IQ)
C
      DO 2 I=1,IQ
      S=0.D0
      DO 1 J=1,N
    1 S=S+DFPDX(I,J)*X(J)
    2 GP(I)=FP(I)-S
      RETURN
      END
      SUBROUTINE FCTX(PRMT,T,X,F,NDIM,U,IR,V,N,V1,V2,IQ)
      IMPLICIT REAL*8 (A-H,O-Z)
      DIMENSION X(NDIM),F(NDIM),U(IR),V(N),V1(N,N),V2(IQ,N),PRMT(5)
      COMMON /Z1/ UT(1,81)
C
      CALL UTIM(T,U,IR,PRMT)
C
      CALL FNLN(T,X,N,L,IR,F,IC)
C
      RETURN
      END
      SUBROUTINE OUTX(PRMT,T,X,F,NDIM,IHLF,AUX,U,IR,V,N,IQ)
      IMPLICIT REAL*8 (A-H,O-Z)
      DIMENSION X(NDIM),F(NDIM),AUX(8,NDIM),U(IR),V(N),PRMT(5)
      COMMON /Z2/ XT(4,81)
C
      C=PRMT(3)
      IT=T/C+1.00002D0
      TE=(IT-1)*PRMT(3)
      IF(DABS(T-TE).GT.1.D-04*C) GO TO 2
      DO 1 I=1,NDIM
    1 XT(I,IT)=X(I)
    2 RETURN
      END
      SUBROUTINE UTIM(T,U,IR,PRMT)
      IMPLICIT REAL*8 (A-H,O-Z)
      DIMENSION U(IR),PRMT(5)
      COMMON /Z1/ UT(1,81)
      B=PRMT(2)
      C=DABS(PRMT(3))
      IF(PRMT(1).GT.PRMT(2)) B=PRMT(1)
      IT=T/C+1.00002D0
      IF(DABS(T-B).LE.1.D-04*C) GO TO 3
      DO 1 I=1,IR
      A1=(UT(I,IT+1)-UT(I,IT))/C
      A0=UT(I,IT)-A1*(IT-1)*C
    1 U(I)=A0+A1*T
    2 RETURN
    3 DO 4 I=1,IR
    4 U(I)=UT(I,IT)
    5 RETURN
      END
      SUBROUTINE XTIM(T,X,N,PRMT)
      IMPLICIT REAL*8 (A-H,O-Z)
      DIMENSION X(N),PRMT(5)
      COMMON /Z2/ XT(4,81)
      B=PRMT(2)
      C=DABS(PRMT(3))
      IF(PRMT(1).GT.PRMT(2)) B=PRMT(1)
      IT=T/C+1.00002D0
      IF(DABS(T-B).LE.1.D-04*C) GO TO 3
      DO 1 I=1,N
      A1=(XT(I,IT+1)-XT(I,IT))/C
      A0=XT(I,IT)-A1*(IT-1)*C
    1 X(I)=A0+A1*T
    2 RETURN
    3 DO 4 I=1,N
    4 X(I)=XT(I,IT)
    5 RETURN
```

```
      END
      SUBROUTINE YPLUS(YP,XP,C,E,IQ,IT,MODE,YP1)
      IMPLICIT REAL*8 (A-H,C-Z)
      DIMENSION YP(IQ),XP(IQ),C(IQ),YP1(IQ)
      YP(IQ)=-1.D0
      IF(IT.LT.2) GO TO 8
      L=IQ-1
      M=IQ-2
      IF(MODE.GT.0) GO TO 5
      IF(L.GT.0) GO TO 2
    1 RETURN
    2 DO 3 I=1,L
    3 YP(I)=YP1(I)+E*(C(I)-XP(I))
    4 RETURN
    5 IF(M.LE.0) GO TO 7
      DO 6 I=1,M
    6 YP(I)=YP1(I)+E*(C(I)-XP(I))
    7 YP(L)=YP1(L)+E*(XP(IQ)/XP(L)-YP(L))
    8 RETURN
      END
      SUBROUTINE FCTP(PRMT,T,P,DP,NDIM,U,IR,X,N,V1,V2,IQ)
      IMPLICIT REAL*8 (A-H,C-Z)
      DIMENSION P(NDIM),DP(NDIM),U(IR),X(N),V1(N,N),V2(IQ,N),PRMT(5)
      COMMON /Z1/ LT(1,81)
      COMMON /Z2/ XT(4,81)
      COMMON /Z3/ B1T(4,4,81)
      COMMON /Z4/ B2T(3,4,81)
C
      CALL UTIM(T,U,IR,PRMT)
C
      CALL XTIM(T,X,N,PRMT)
C
      CALL DFDXT(T,X,N,U,IR,V1)
C
      CALL DFPDXT(T,X,N,U,IR,V2,IQ)
C
      L=N+IQ
      DO 2 I=1,L
      DO 2 J=1,N
      S=0.D0
      DO 1 K=1,N
    1 S=S-P(I+(K-1)*L)*V1(K,J)
    2 DP(I+(J-1)*L)=S
      M=N+1
      DO 3 I=M,L
      DO 3 J=1,N
      K=I+(J-1)*L
    3 DP(K)=DP(K)-V2(I-N,J)
    4 RETURN
      END
      SUBROUTINE OUTP(PRMT,T,P,DP,NDIM,IHLF,AUXP,U,IR,X,N,IQ)
      IMPLICIT REAL*8 (A-H,C-Z)
      DIMENSION P(NDIM),DP(NDIM),AUXP(8,NDIM),U(IR),X(N),PRMT(5)
      COMMON /Z3/ B1T(4,4,81)
      COMMON /Z4/ B2T(3,4,81)
C
      C=DABS(PRMT(3))
      IT=T/C+1.00002D0
      TE=(IT-1)*C
      IF(DABS(T-TE).GT.1.D-04*C) GO TO 3
      L=N+IQ
      DO 1 I=1,N
      DO 1 J=1,N
    1 B1T(I,J,IT)=P(I+(J-1)*L)
      M=N+1
      DO 2 I=M,L
      DO 2 J=1,N
    2 B2T(I-N,J,IT)=P(I+(J-1)*L)
    3 RETURN
      END
      SUBROUTINE FCTXL(T,X,N,U,IR,FL,IQ)
      IMPLICIT REAL*8 (A-H,C-Z)
      DIMENSION X(N),U(IR),FL(N),DFDX(4,4),G(4),UMAX(4)
      COMMON /Z3/ B1T(4,4,81)
      COMMON /Z5/ IT
C
      DO 4 I=1,IR
    4 UMAX(I)=U(I)
      CALL GLIN(T,X,N,U,UMAX,IR,FL,DFDX,G,IQ)
      DO 2 I=1,N
      S=0.D0
      DO 1 J=1,N
    1 S=S+B1T(I,J,IT)*G(J)
    2 FL(I)=S
    3 RETURN
      END
      SUBROUTINE RALIN(PRMT,X,N,XP,IQ,C,D,XJ,XBJ,NP,IR,MODE,ALFA,IT)
      IMPLICIT REAL*8 (A-H,C-Z)
      DIMENSION X(N),XP(IQ),C(IQ),PRMT(5)
      COMMON /Z1/ LT(1,81)
      COMMON /Z2/ XT(4,81)
  100 FORMAT(1H ,75(1H+)/1H ,'RESULTS OF ITERATION',I4,2X,'ALFA=',
     1D13.6//)
  101 FORMAT(1H ,'PERIODIC END STATE X1 ... X4'/1H ,4(D13.6,2X)//)
  102 FORMAT(1H ,'OBJECTIVE FUNCTION=',D13.6/)
  103 FORMAT(1H ,'DISTANCE FROM THE DESIRED CONSTRAINTS=',D13.6/)
  104 FORMAT(1H ,'AUGMENTED OBJECTIVE FUNCTION=',D13.6//)
  105 FORMAT(1H ,4X,'T',13X,'U',13X,'X1',12X,'X2',12X,'X3',12X,'X4'//
  106 FORMAT(1H ,7(D13.6,2X))
  107 FORMAT(1H ,2X//1H ,75(1H+)/)
```

```
      C
      WRITE(6,100) IT,ALFA
      WRITE(6,101) (X(I),I=1,N)
      WRITE(6,102) XJ
      WRITE(6,103) D
      WRITE(6,104) XBJ
      WRITE(6,105)
      DO 1 I=1,NP
      T=(I-1)*DABS(PRMT(3))
    1 WRITE(6,106) T,(UT(J,I),J=1,IR),(XT(J,I),J=1,N)
      WRITE(6,107)
    2 RETURN
      END
      SUBROUTINE RLIN(PRMT,XS,N,XP,YP,C,IC,D,XJ,XBJ,NP,IR,E,ITL,ITST)
      IMPLICIT REAL*8 (A-H,O-Z)
      DIMENSION PRMT(5),XS(N),XP(IC),YP(IC),C(IC)
      COMMON /Z1/ UT(1,81)
      COMMON /Z2/ XT(4,81)
  100 FORMAT(1H ,30(1H.)//1H ,'ITL=',I4,2X,'E=',D13.6,2X,'ITST=',I2/)
  102 FORMAT(1H ,'CONSTRAINTS ARE XP1 ... '/1H ,4(D13.6,2X)/)
  101 FORMAT(1H ,'ADJOINT CONSTANTS ARE YP1 ...'/1H ,4(D13.6,2X)/)
  103 FORMAT(1H ,'JL=',D13.6,2X,'D=',D13.6,2X,'JBL=',D13.6/)
  104 FORMAT(1H ,'OPTIMAL CONTROL FOR LINEAR SYSTEM IS:'//)
  105 FORMAT(1H ,4(D13.6,2X))
  106 FORMAT(1H ,2X//1H ,30(1H.)/)
    1 WRITE(6,100) ITL,E,ITST
      WRITE(6,101) (YP(I),I=1,IC)
      WRITE(6,102) (XP(I),I=1,IC)
      WRITE(6,103) XJ,D,XBJ
      IF(ITL.GE.0) RETURN
      WRITE(6,104)
      DO 2 J=1,NP
      T=(J-1)*DABS(PRMT(3))
    2 WRITE(6,105) T,(UT(I,J),I=1,IR)
      WRITE(6,107) (XS(I),I=1,N)
  107 FORMAT(1H /1H ,'OPTIMUM PERIODIC END STATE X1...'/1H ,4(D13.6,2X))
    3 WRITE(6,106)
    4 RETURN
      END
      SUBROUTINE DRKGS(PRMT,Y,DERY,NDIM,IHLF,FCT,OUT,AUX,U,IR,V,N,V1,V2,
     1IC)
      IMPLICIT REAL*8 (A-H,C-Z)
      DIMENSION Y(NDIM),DERY(NDIM),AUX(8,NDIM),PRMT(5),A(4),B(4),C(4)
      DIMENSION U(IR),V(N),V1(IC,N),V2(IC,N)
      COMMON /Z1/ UT(1,81)
      COMMON /Z2/ XT(4,81)
      COMMON /Z3/ B1T(4,4,81)
      COMMON /Z4/ B2T(3,4,81)
      DO 1 I=1,NDIM
    1 AUX(8,I)=DERY(I)/15.0D0
      X=PRMT(1)
      XEND=PRMT(2)
      H=PRMT(3)
      PRMT(5)=0.0D0
      CALL FCT(PRMT,X,Y,DERY,NDIM,U,IR,V,N,V1,V2,IC)
      IF(H*(XEND-X)) 38,37,2
    2 A(1)=0.5D0
      A(2)=1.0D00-DSQRT(2.0D00)/2.0D00
      A(3)=1.0D00+DSQRT(2.0D00)/2.0D00
      A(4)=1.0D0/6.0D0
      B(1)=2.0D0
      B(2)=1.0D0
      B(3)=1.0D0
      B(4)=2.0D0
      C(1)=0.5D0
      C(2)=A(2)
      C(3)=A(3)
      C(4)=0.5D0
      DO 3 I=1,NDIM
      AUX(1,I)=Y(I)
      AUX(2,I)=DERY(I)
      AUX(3,I)=0.0D0
    3 AUX(6,I)=0.0D0
      IREC=0
      H=H+H
      IHLF=-1
      ISTEP=0
      IEND=0
    4 IF((X+H-XEND)*H) 7,6,5
    5 H=XEND-X
    6 IEND=1
    7 CALL OUT(PRMT,X,Y,DERY,NDIM,IHLF,AUX,U,IR,V,N,IQ)
      IF(PRMT(5)) 40,8,40
    8 ITEST=0
    9 ISTEP=ISTEP+1
      J=1
   10 AJ=A(J)
      BJ=B(J)
      CJ=C(J)
      DO 11 I=1,NDIM
      R1=H*DERY(I)
      R2=AJ*(R1-BJ*AUX(6,I))
      Y(I)=Y(I)+R2
      R2=R2+R2+R2
   11 AUX(6,I)=AUX(6,I)+R2-CJ*R1
      IF(J-4) 12,15,15
   12 J=J+1
      IF(J-3) 13,14,13
   13 X=X+0.5D0*H
   14 CALL FCT(PRMT,X,Y,DERY,NDIM,U,IR,V,N,V1,V2,IQ)
```

```
      GO TO 10
   15 IF(ITEST) 16,16,20
   16 DO 17 I=1,NDIM
   17 AUX(4,I)=Y(I)
      ITEST=1
      ISTEP=ISTEP+ISTEP-2
   18 IHLF=IHLF+1
      X=X-H
      H=0.5D0*H
      DO 19 I=1,NDIM
      Y(I)=AUX(1,I)
      DERY(I)=AUX(2,I)
   19 AUX(6,I)=AUX(3,I)
      GO TO 9
   20 IMOD=ISTEP/2
      IF(ISTEP-IMOD-IMOD) 21,23,21
   21 CALL FCT(PRMT,X,Y,DERY,NDIM,U,IR,V,N,V1,V2,IQ)
      DO 22 I=1,NDIM
      AUX(5,I)=Y(I)
   22 AUX(7,I)=DERY(I)
      GO TO 9
   23 DELT=0.0D0
      DO 24 I=1,NDIM
   24 DELT=DELT+AUX(8,I)*DABS(AUX(4,I)-Y(I))
      IF(DELT-PRMT(4)) 28,28,25
   25 IF(IHLF-10) 26,36,36
   26 DO 27 I=1,NDIM
   27 AUX(4,I)=AUX(5,I)
      ISTEP=ISTEP+ISTEP-4
      X=X-H
      IEND=0
      GO TO 18
   28 CALL FCT(PRMT,X,Y,DERY,NDIM,U,IR,V,N,V1,V2,IQ)
      DO 29 I=1,NDIM
      AUX(1,I)=Y(I)
      AUX(2,I)=DERY(I)
      AUX(3,I)=AUX(6,I)
      Y(I)=AUX(5,I)
   29 DERY(I)=AUX(7,I)
      CALL OUT(PRMT,X-H,Y,DERY,NDIM,IHLF,AUX,U,IR,V,N,IQ)
      IF(PRMT(5)) 40,30,40
   30 DO 31 I=1,NDIM
      Y(I)=AUX(1,I)
   31 DERY(I)=AUX(2,I)
      IREC=IHLF
      IF(IEND) 32,32,35
   32 IHLF=IHLF-1
      ISTEP=ISTEP/2
      H=H+H
      IF(IHLF) 4,33,33
   33 IMOD=ISTEP/2
      IF(ISTEP-IMOD-IMOD) 4,34,4
   34 IF(DELT-0.02D0*PRMT(4)) 35,35,4
   35 IHLF=IHLF-1
      ISTEP=ISTEP/2
      H=H+H
      GO TO 4
   36 IHLF=11
      CALL FCT(PRMT,X,Y,DERY,NDIM,U,IR,V,N,V1,V2,IQ)
      GO TO 39
   37 IHLF=12
      GO TO 39
   38 IHLF=13
   39 CALL OUT(PRMT,X,Y,DERY,NDIM,IHLF,AUX,U,IR,V,N,IQ)
   40 CONTINUE
      RETURN
      END
      SUBROUTINE INVERT(P10,N,V1)
      IMPLICIT REAL*8 (A-H,O-Z)
      DIMENSION P10(N,N),V1(N,N)
      DIMENSION DUM(4)
      DO 2 I=1,N
      DO 1 J=1,N
    1 V1(I,J)=-P10(I,J)
    2 V1(I,I)=1.D0+V1(I,I)
C
      CALL MATIN(N,V1,1.D-20,DETER)
C
      RETURN
      END
      SUBROUTINE MATIN(N,A,EPS,D)
      IMPLICIT REAL*8 (A-H,O-Z)
      DIMENSION A(N,N),IR(4),JC(4),JO(4),Y(4)
      D=1.D0
      DO 7 K=1,N
      KM1=K-1
      PIVOT=0.D0
      DO 3 I=1,N
      DO 3 J=1,N
      IF(K.EQ.1) GO TO 2
      DO 1 IS=1,KM1
      DO 1 JS=1,KM1
      IF(I.EQ.IR(IS)) GO TO 3
      IF(J.EQ.JC(JS)) GO TO 3
    1 CONTINUE
    2 IF(DABS(A(I,J)).LE.DABS(PIVOT)) GO TO 3
      PIVOT=A(I,J)
      IR(K)=I
      JC(K)=J
    3 CONTINUE
```

```
      IF(DABS(PIVOT).GT.EPS) GO TO 4
      D=0.D0
      WRITE(6,100)
      RETURN
    4 IRK=IR(K)
      JCK=JC(K)
      D=D*PIVOT
      DO 5 J=1,N
    5 A(IRK,J)=A(IRK,J)/PIVOT
      A(IRK,JCK)=1.D0/PIVOT
      DO 7 I=1,N
      AIJ=A(I,JCK)
      IF(I.EQ.IRK) GO TO 7
      A(I,JCK)=-AIJ/PIVOT
      DO 6 J=1,N
    6 IF(J.NE.JCK) A(I,J)=A(I,J)-AIJ*A(IRK,J)
    7 CONTINUE
      DO 8 I=1,N
      IRI=IR(I)
      JCI=JC(I)
    8 JO(I)=JCI
      INT=0
      NM1=N-1
      DO 9 I=1,NM1
      IP1=I+1
      DO 9 J=IP1,N
      IF(JC(J).GE.JO(I)) GO TO 9
      JT=JC(J)
      JC(J)=JC(I)
      JC(I)=JT
      INT=INT+1
    9 CONTINUE
      IF(INT/2*2.NE.INT) D=-D
      DO 11 J=1,N
      DO 10 I=1,N
      IRI=IR(I)
      JCI=JC(I)
   10 Y(JCI)=A(IRI,J)
      DO 11 I=1,N
   11 A(I,J)=Y(I)
      DO 13 I=1,N
      DO 12 J=1,N
      IRJ=IR(J)
      JCJ=JC(J)
   12 Y(IRJ)=A(I,JCJ)
      DO 13 J=1,N
   13 A(I,J)=Y(J)
  100 FORMAT(1H ,'MAXIMUM PIVOT LESS THAN EPS=1.D-20')
   14 RETURN
      END
```

```

.SAMPLE DATA FOR THE PROGRAM

 +0.100C000CD C1+1.2500CC00D-02+1.000C0000D-08
      1     1
 +0.1C0C00CCD C1+0.1CCCCC00D 01
 +0.1C0C00CCD C2
      4     1
```